

# Effect of sludge-processing mode, soil texture and soil pH on metal mobility in undisturbed soil columns under accelerated loading

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## Abstract

The effect of sludge processing (digested dewatered, pelletized, alkaline-stabilized, composted, and incinerated), soil type and initial soil pH on trace metal mobility was examined using undisturbed soil columns. Soils tested were Hudson silt loam (Glossaquic Hapludalf) and Arkport fine sandy loam (Lamellic Hapludalf), at initial pH levels of 5 and 7. Sludges were applied during four accelerated cropping cycles (215 tons/ha cumulative application for dewatered sludge; equivalent rates for other sludges), followed by four post-application cycles. Also examined (with no sludge applications) were Hudson soil columns from a field site that received a heavy loading of sludge in 1978. Romaine (*Lactuca sativa*) and oats (*Avena sativa*) were planted in alternate cycles, with oats later replaced by red clover (*Trifolium pratense*). Soil columns were watered with synthetic acid rainwater, and percolates were analyzed for trace metals (ICP spectroscopy), electrical conductivity and pH. Percolate metal concentrations varied with sludge and soil treatments. Composted sludge and ash had the lowest overall metal mobilities. Dewatered and pelletized sludge had notable leaching of Ni, Cd and Zn in Arkport soils, especially at low pH. Alkaline-stabilized sludge had the widest range of percolate metals (relatively insensitive to soils) including Cu, Ni, B and Mo. Old site column percolate concentrations showed good agreement with previous field data. Little leaching of P was observed in all cases. Cumulative percolate metal losses for all treatments were low relative to total applied metals. Leachate and soil pH were substantially depressed in dewatered and pelletized sludge soil columns and increased for alkaline-stabilized and ash treatments. © 2000 Elsevier Science Ltd. All rights reserved.

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## 1. Introduction

Reuse of municipal wastewater sludge via land application recycles the organic matter (which improves soil physical characteristics) and nutrients in the sludge. Reuse is, however, complicated by the low but still significant levels of contaminants present in the sludge. Of these, trace metals have received the most attention to date. The risks of human, crop and/or environmental toxicity posed by these elements are a function of their mobility and availability.

Sludges can be processed by a variety of methods to reduce sludge mass, volume, odors and/or pathogen

viability. In an earlier article (Richards et al., 1997) we showed that the mode (drying, composting, alkaline stabilization, or incineration) by which dewatered sludge was processed significantly affected not only trace element concentrations but also their in vitro leachability, as determined by the Toxicity Characteristic Leaching Procedure (TCLP; USEPA, 1992a). Using these same sludge products, Theis et al. (1998) found metal concentrations in leachate from these products followed the pattern of: alkaline-stabilized > dried pellets > dewatered sludge > incinerated ash > composted. Attention has been given to the effects of processing mode on availability of N (Misselbrooke et al., 1996; Shepherd, 1996) and P (Frossard et al., 1996; Wen et al., 1997), as also summarized in the recent reviews by Krogman et al. (1997, 1998).

Soil pH and soil texture play important roles in controlling trace metal mobility, with most metals (in free

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ionic form) being most mobile in acidic, coarse-textured soils (McBride, 1994). Solubility and plant uptake of Cd and Zn were greater from a non-limed sludge than from a lime-stabilized sludge (Basta and Sloan, 1999). Acid forest soils with lower total Cd concentrations than agricultural nevertheless had far greater soluble Cd concentrations due to lower pH levels (Römken and Salomons, 1998). Mobility can, however, also be significant at circumneutral or higher pH due to metal complexation with dissolved organic matter (DOM) which itself becomes more soluble at those pH levels. As a result, alkaline-stabilized sludge products have been shown to have TCLP extractabilities of 25–50% of total Cu, Ni and Mo (Richards et al., 1997), with similar results for water extractabilities (McBride, 1998). Organic and inorganic colloids have been shown to accelerate the subsurface mobility of many contaminants (McCarthy and Zachara, 1989) particularly where DOM levels are elevated and contaminants have a high affinity for the mobile colloids. Xiao et al. (1999) reported ash/sludge mixtures as having elevated DOM concentrations that increased trace metal leachability, and Jordan et al. (1997) found increases in Pb solubility in the presence of DOM. Lamy et al. (1993) observed DOM facilitation of Cd mobility following sludge application.

Substantial deficits of applied sludge-borne metals are apparent for many field studies reporting mass balances (or when balances are performed on reported data). These studies are summarized by McBride et al. (1997) and Richards et al. (1998). More recently, Baveye et al. (1999) concluded that from 36 to 60% of applied metals were lost in the experimental sludge application plots of Hinesly et al. (1984), even when total soil dissolution was employed to ensure soil metal recovery. Tillage dispersion or incomplete analytical recovery may account for some of the shortfall in applied metals in some cases (McGrath and Lane, 1989; Chang et al., 1984). These factors are not applicable in all cases, and researchers, assuming soil metal immobility, are often forced to conclude that reported applications were incorrect (Baxter et al., 1983; Streck and Richter, 1997). Leaching losses of metals have been cited as a potential (if unlikely) mechanism of loss (McGrath and Lane, 1989; Dowdy et al., 1991). Leaching losses are often ruled out due to lack of observable increases in subsoil metals concentrations (Baxter et al., 1983), but we have recently demonstrated that metal leaching is not necessarily accompanied by detectable subsoil readsorption within 1.5 m depth (Richards et al., 1998). Barbarick et al. (1998) did detect increases in subsoil Zn despite limited soil moisture regime (dryland wheat), and Brown et al. (1997) noted subsoil increases in several metals.

Duncomb et al. (1982) reported little significant increase in soil solution metal concentrations at depths of 60 and 150 cm following repeated sludge applications. Jackson et al. (1999) reported little increases in

soil solution concentrations at 10 cm depth from sludge/ash applications. However, these and other studies used ceramic cup lysimeters for water sampling which have been shown to absorb trace metals from samples (McGuire et al., 1992; Wenzel et al., 1997). Preferential flow paths in the soil are also likely to be missed by suction cup lysimeters (Boll, 1995), or may be altered by installation procedures such as packing with slurried soil (Jackson et al., 1999).

USEPA (1992b) predicted very limited potential for leaching of sludge-borne trace metals, but the risk assessment utilized a very narrow data base, and was based on modeling approaches that excluded organic-facilitated transport and that assumed conventional uniform flow through homogenous soil and aquifer strata. Preferential flow through soil macropores or via fingering phenomena has been shown to result in greater mobilities (Kung, 1990; Steenhuis et al., 1995, 1996) than would be predicted by conventional uniform flow models for a range of contaminants. Camobreco et al. (1996) reported that conventionally packed soil columns (which force uniform water flow) were overly optimistic about soil metal retention capacity when compared to more realistic undisturbed soil columns that preserve preferential flow paths. In contrast, most soil column studies reporting metal immobility utilized conventional packed soil columns (Welch and Lund, 1987).

The goal of the present study was to use 90 undisturbed soil columns to determine the effects of sludge-processing mode, initial soil pH and soil texture on the short- and long-term mobility of metals and nutrients. The sludge products (detailed in Richards et al., 1997) used in the study were all derived from the same sludge feedstock to allow valid comparison of processing effects. This article reports observed percolate pH, conductivity and soluble metals concentrations as well as soil pH trends.

## 2. Experimental approach

The primary experiment (Table 1) examined two soils (coarse vs. fine textured) with no prior history of sludge application. Five sludge products—consisting of de-watered digested sludge and four sludge products derived from it via composting, alkaline stabilization, drying and pelletization and incineration—were applied to the soils. Initial soil pH levels were adjusted to low (pH 5) and circumneutral (pH 6.5–7) levels. No-sludge controls were operated at low and neutral pH levels, and additional ‘natural control’ columns were operated with no pH adjustments or nutrient additions to provide an absolute ‘no additions’ baseline. All treatments were examined using triplicate columns.

A third soil, an ‘old site’ fine-textured soil with a history of sludge application, was used for a series of

Table 1  
Controlled application soil column study experimental matrix, showing number of columns assigned to each treatment of sludge and pH

Sludge and pH treatments		Soil type		
Sludge type	Initial soil pH	Arkport sandy loam	Hudson silt loam	Old site Hudson
1. Digested dewatered	5	3	3	–
	7	3	3	–
2. Composted	5	3	3	–
	7	3	3	–
3. Alkaline-stabilized (N-Viro)	5	3	3	–
	7	3	3	–
4. Dried and pelletized	5	3	3	–
	7	3	3	–
5. Incinerated ash	5	3	3	–
	7	3	3	–
6. Control	5	3	3	3
	7	3	3	3
	Natural	3	3	3
	7+	–	–	3
Total of each soil type		39	39	12
Total soil columns			90	

controls at low, neutral, natural and high (>7) pH levels. No additional sludge was applied to these columns. The columns were used to: (1) compare column leachate results with those from in situ passive wick lysimeters installed in the original field plots; and (2) observe the effects of altering soil pH on residual metals present in the soil.

In all cases, undisturbed soil columns were used to better simulate field soil conditions by preserving natural preferential flow paths. Accelerated cropping and leaching cycles were used, with sufficient simulated acid rain applied during each 3-month cropping cycle to result in a calendar year's volume of percolate.

### 2.1. Source soil descriptions

Soil columns were extracted in the summer of 1993 from college farmland adjacent to the Cornell campus in Ithaca, NY. All soils had similar elevation and slope aspect (level or slight northward slope), and all were essentially free of rocks or gravel, simplifying both field extraction and management in the greenhouse. All sites were downwind and within approximately 1 km of the coal-fired University steam plant.

The fine-textured soil was Hudson silt loam (fine, illitic, mesic, Glossaquic Hapludalf), thought to be lacustrine in origin, with a silt loam epipedon (surface horizon) underlain by a silty clay loam subsoil. Mean horizon depths were A<sub>p</sub> 15 cm, E 25 cm and BE to column depth. Soil cores were excavated from a field

used as unimproved pasture for at least the past 25 years. The coarse-textured soil was an Arkport fine sandy loam (coarse loamy, mixed, mesic, active, Lamellic Hapludalf), presumably a small deltaic deposit. The sandy loam topsoil (A1 to 12 cm mean depth, A2 to 25 cm) was underlain by a variety of subsoil horizons: fine sand, loamy sand and silty sand. The Arkport area was about 0.3 km from the Hudson site, and was similarly used as long-term unimproved pasture. Thirty-nine cores were taken from each of these sites.

The old site soil columns were excavated from an experimental sludge application plot in the Cornell Orchards, on Hudson silt loam soils that were in fact contiguous with the pasture from which the other Hudson columns were taken. Sludge was applied to the plot (previously an old apple orchard) in 1978 in a single heavy loading (244 tons/ha nominal rate). Following several years of experimental row cropping, the site was plowed and dwarf apple trees were planted in 1986. Site history and soil characteristics are discussed in greater detail elsewhere (McBride et al., 1997; Richards et al., 1998). Mean horizon depths were A<sub>p</sub> 25 cm (with inclusions of blocks of B resulting from deep tillage), B1 to 30 cm and B2 to column depth. Wick lysimeters were installed in 1993 to monitor percolate metal concentrations as reported in Richards et al. (1998). Twelve soil cores were concurrently extracted from the perimeter of the excavation pit dug for installation of the wick lysimeters.

## 2.2. Soil columns

Whereas the old site columns were dug from the periphery of the wick lysimeter pit in the Orchards sludge plot, column extraction of the other two soil types was facilitated by the use of a backhoe to excavate long trenches. Columns were then hand-excavated along the edges of these trenches. A column of soil (28 cm diameter and 35 cm deep) was exposed by carefully excavating surrounding soil. The soil profile of each column was described in the field, and soil samples from the periphery of each column were taken in accordance with the horization. A 35 cm length of 30-cm ID corrugated black polyethylene culvert was placed over the column, and minimal-expansion foam (commercially available “Great Stuff” polyurethane) was injected into the gap between the soil column and culvert and allowed to cure overnight. The column was then removed by digging under the column. Excess soil was removed from the base of the column, and the base was carefully ‘picked’ to remove any smeared soil to ensure that flow paths would be intact.

Each column was placed on a support base (Fig. 1), with a central drain hole. The column rested on two 1.2-m diameter circles of black polyethylene film, which were drawn up and secured around the column. A circle of foam padding (2 cm thick) under the black plastic ensured contact between the plastic and the base soil. To direct leachate towards the central drain hole, a

ridge of 1.3-cm thick foam weatherstripping was placed around the outer edge of the foam base, and radial notches were cut into the foam base. PVC fittings threaded together through the drain hole both secured the plastic film to the base and provided a water-tight seal. Leachate was directed through plastic tubing connected to the elbow to a polyethylene storage jug, with both tubing and jug darkened to retard algal growth.

Individual reservoirs (3.3 l volume) were filled weekly to dispense water to each soil column. The water applied for each cropping cycle was designed to result in approximately 30 cm depth of percolate, the typical recharge rate for this area. In order to moderate the rate of inflow to each column, each reservoir was fitted with a constant-head device and a short piece of narrow diameter tubing to serve as an in-line flow restrictor. A network of short fiberglass wicks was used to distribute the flow evenly across the soil surface of each column. Synthetic acid rain was used (Table 2; sulfate was inadvertently 20% lower than 4.96 mg/l target), prepared each week by diluting a 10000× concentrate with de-ionized water. A 500-l polyethylene central mixing tank and pump were used for mixing and distributing the water to the column reservoirs.

Column extraction records and soil profiles were examined to determine the variability of soil characteristics between columns. This was done to assure that column variabilities were equally represented in the various treatments to be examined. For the 39 Hudson soil columns there were no notable differences between columns other than a normal variation in horizon depths. Replicates were assigned on the basis of location within the field (one replicate each from middle, left and right sides of the excavation area). The 39 coarse-textured Arkport soil columns were similarly assigned on the basis of field location. Being a deltaic deposit, variation of subsoil characteristics was more marked across the field. However, assignment on the basis of field location well distributed this variation. Columns in one end of the field (assigned to the first replicate of each treatment) generally had a thin silty subsoil horizon

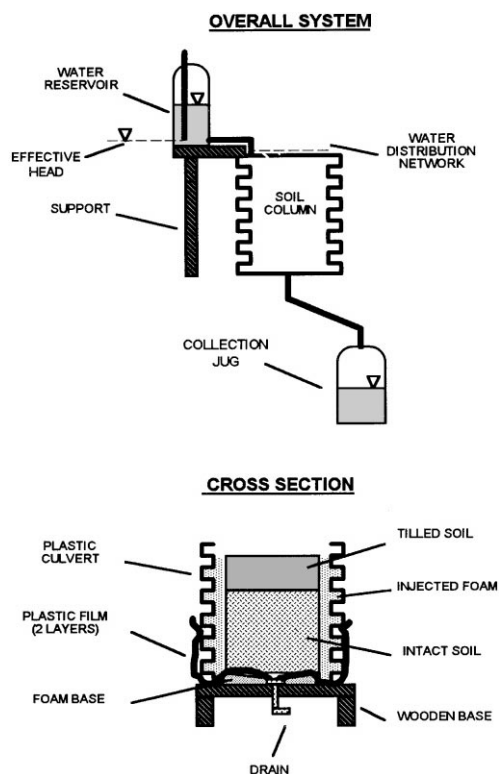


Fig. 1. Soil column system and column cross-section.

Table 2

Artificial rainwater ionic composition (T.L. Theis, 1993, personal communication)<sup>a</sup>

Ion	Conc. (mg/l)
Na <sup>+</sup>	0.15
NH <sub>4</sub> <sup>+</sup>	0.32
K <sup>+</sup>	0.09
SO <sub>4</sub> <sup>2-</sup>	3.96 <sup>b</sup>
Ca <sup>2+</sup>	0.83
NO <sub>3</sub> <sup>-</sup>	2.88
Mg <sup>2+</sup>	0.08
Cl <sup>-</sup>	0.47

<sup>a</sup> Approximate pH 4–4.5.

<sup>b</sup> Sulfate inadvertently lower than 4.96 mg/l target concentration.

absent in the other two replicates. The 12 columns extracted from the Cornell Orchards old site were grouped into three categories: (1) columns with visible dark veins of organic matter in the  $A_p$  horizon due to incomplete tillage of sludge when applied; (2) columns with thin B1 horizons; and (3) all other columns. One column from each of these three categories was assigned to each treatment so that any effects due to initial column conditions would be evenly represented in each treatment.

Columns were stored indoors, sparingly watered to prevent desiccation, and covered with black plastic to kill weeds. Columns were placed in the greenhouse in summer of 1994. To prevent effects due to location within the greenhouse (which had a crossflow ventilation pattern), the greenhouse was divided into three areas, with one replicate of each treatment assigned to each area. Column locations within each replicate's designated area in the greenhouse were randomly determined. The upper 10 cm of soil was carefully hand-tilled in preparation for pH adjustment and sludge incorporation. Once flow systems were installed and hand-tilling was complete, several initial leachings with synthetic acid rain were performed.

In August 1994, additions of lime (reagent grade  $\text{CaCO}_3$ ) or acid (0.5 N  $\text{H}_2\text{SO}_4$ ) were made to adjust soil pH levels of the upper 10 cm to target initial levels. Additions were made incrementally and iteratively over a period of several weeks, based on lime requirement and acid titration analyses and resulting soil pH levels. Cumulative lime additions (g  $\text{CaCO}_3$ /column) for columns assigned to an initial pH of 7 were 26.8 (Hudson), 24.8 (Arkport) and 55.0 (old site Hudson). Addition rates for high pH old site Hudson columns were 182.5 g/column. Acid additions (meq/column) for columns assigned to an initial pH of 5 were 435 meq (old site Hudson) and 138 meq (Hudson). Initial pH levels of Arkport soils were sufficiently low so that no acid additions were necessary for low pH conditions. Following pH adjustment, three more leachings were carried out.

Prior to cropping cycle 8, columns in pH 7 treatments were restored to near pre-Cycle 1 pH levels by lime additions, while low pH treatments were not adjusted in order to simulate unmanaged conditions. Lime addition rates for pH 7 pellets, compost and control columns were 26.8 (Hudson) and 24.8 (Arkport) g/column. For pH 7 dewatered sludge treatments, addition rates were 53.6 (Hudson) and 49.6 (Arkport) g/column. Additions for old site Hudson high pH columns were 182.5 g/column. No additions were needed for N-Viro or ash columns.

### 2.3. Sludge characteristics

Historically, comparisons of different sludge products are weakened by the fact that the sludge feedstock for each process differs in composition. A significant effort

(coordinated by the New York State Energy Research and Development Authority) was thus made to ensure direct comparability of the various sludge processes by producing all products from the same sludge feedstock. The sludge products used were thus all derived from dewatered digested sludge produced during a single day (16 May 1994) at the Onondaga County wastewater treatment facility in Syracuse, NY. The dewatered digested sludge (DW) produced at the plant was the feedstock for the other processes and was itself used in the study. Composted sludge (COM) was obtained by shipping 30 tons of the dewatered sludge to Lockport, NY, where it was mixed with virgin wood chips, composted and cured for several months in a municipal composting facility. Dried sludge pellets (PELL) were obtained by pelletizing and drying several hundred kilograms of sludge in a pilot-scale mill at Clarkson University (Potsdam, NY). Incinerated sludge ash (ASH) was produced by incinerating over 50 metric tons (wet wt.) in a multiple hearth furnace at Monroe County's Northwest Quadrant facility (Rochester, NY). Alkaline-stabilized sludge (NV; N-Viro™ process) was obtained from the Waste Stream Environmental facility at the Onondaga County wastewater plant. Detailed processing information and analyses, including TCLP extractability, have been summarized elsewhere (Richards et al., 1997).

Sludge composition and cumulative loadings are summarized in Table 3. Application rates of the various sludge products were normalized to the amount of dewatered sludge dry matter initially present in each process, with the goal being equal loading rates of sludge-derived metals. Normalization factors (g product TS per g initial DW TS) were based on total solids for pellets, nonvolatile solids for ash, reported amendment ratios for N-Viro and reported wood chip additions and estimated biodegradation for compost.

A three-phase sludge loading program was followed (Table 4). During Phase 1, columns were given agronomic (i.e. typical N-based) sludge loadings of 7.5 tons/ha (DW sludge-equivalent) per cycle for two application/cropping cycles (Cycles 1 and 2). The only exception was that the Cycle 2 N-Viro applications for high pH columns were deferred to and added to the Cycle 3 application. Phase 2 consisted of two heavy loading cycles (Cycles 3 and 4) of 100 tons/ha DW sludge each, to rapidly attain cumulative metals loading in the soil to simulate long-term applications. This phase allowed rapid attainment of a cumulative metals content in soil equivalent to 28 years at the 7.5 tons/ha rate (cumulative DW sludge loading rate of 215 tons/ha). Although these heavy loading rates were obviously much higher than agronomic rates, they were still in the range of single-application loadings used for land reclamation. During Phase 3 no additional sludge was applied, but cropping and leaching cycles were continued to observe long-term post-application effects.

Table 3  
Sludge product cumulative total solids and elemental loadings per column

Sludge product	Dewatered	Pellets	Composted	N-Viro	Ash
<i>Sludge loading</i>					
Normalization factor	1	1	1.1	3	0.45
Dry matter (g/column)	1300	1352	1524	4064	599
Dry matter (tons/ha)	212	221	249	663	98
<i>Metals loadings (kg/ha)</i>					
Ca	9020	8360	9670	215 620	10 290
Cd	1.19	1.05	1.42	1.05	0.35
Cr	27.6	30.2	29.7	26.7	21.3
Cu	124	117	134	79	119
Fe	14 390	12 950	15 330	9570	11 230
K	255	457	261	1450	416
Mg	1270	1330	1340	7990	1660
Mn	72.0	120.3	77.5	162.4	81.3
Mo	6.13	4.73	7.09	3.78	5.39
Na	155	135	163	228	213
Ni	7.59	8.08	8.38	8.41	7.30
P	5700	5130	6110	3240	7020
Pb	28.0	27.1	30.1	NA <sup>a</sup>	14.1
S	3360	2450	3430	5610	1040
Zn	116	114	125	76	94

<sup>a</sup> Direct analysis not available due to spectral interference. Estimated rate 28–30 kg/ha.

Table 4  
Undisturbed soil column system: operation summary<sup>a</sup>

Cycle	Dates	Weekly waterings	Loading rate tons/ha (DW sludge)	Crop	Total nutrients added (number of equal additions in brackets)
0	7/94–10/94	4	none (pre-application)	None	None
1	11/94–2/95	15	7.5	Oats	ASH, CTRL: 40 kgN/ha NV, COM: 19 kgN/ha (1)
2	4/95–7/95	16	7.5	Romaine	ASH, CTRL: 120 kgN/ha PELL: 63 kgN/ha COM, NV: 100 kgN/ha (5)
3	9/95–12/95	13	100	Oats	ASH, CTRL: 40 kgN/ha (1)
4	1/96–4/96	12	100	Romaine	80 kgN/ha (ASH, CTRL) (2)
5	5/96–8/96	12	0	Oats	None
6	1/97–3/97	12	0	Romaine	80 kgN/ha (ASH, CTRL) (2) 80 kgK/ha (all but NCTRL) (1)
7	10/97–1/98	16	0	Red clover	None
8	4/98–7/98	12	0	Romaine	None

<sup>a</sup> DW, dewatered digested sludge; ASH, incinerated sludge ash; CTRL, control; NV, alkaline-stabilized sludge; COM, composted sludge; PELL, dried sludge pellets.

Sludge was added to the mixed topsoil layer (previously hand-tilled to 10 cm depth) at the beginning of each application/cropping cycle (Cycles 1–4). The mixed layer was carefully excavated to the original 10 cm depth and mixed in a polyethylene tub. A soil sample was then taken, preweighed masses of sludge were added and the soil and sludge were thoroughly mixed. The soil/sludge mixture was then returned to the soil column and firmly pressed into place. Any large roots or plant residues in the columns were placed on top of the exposed subsoil in the column prior to returning the soil. The same excavation and mixing procedure was used to obtain soil samples in subsequent post-application cropping cycles.

#### 2.4. Crops and watering

Crops were grown on the soil columns to: (1) provide an index of phytoavailability and/or phytotoxicity via crop response (to be reported in subsequent publications); (2) better simulate field conditions by maintaining an active rhizosphere in the soil and allowing root growth to open and maintain preferential flow paths; and (3) provide a more realistic pattern of soil moisture content and percolation rates over the cropping cycle (percolation during early growth and after harvest but little or no percolate during mid-cycle). Relatively short-season, shallow-rooted crops were grown in alternate cropping cycles (Table 4). Oats (*Avena sativa* var. Ogle; used in Cycles 1, 3 and 5) represent a field crop that is

relatively indifferent to trace metals in terms of uptake and/or phytotoxicity. In Cycle 7 and following, oats were replaced by red clover (*Trifolium pratense*), a common hay/forage crop that exhibits a degree of sensitivity to soil metals. Romaine (or Cos) lettuce (*Lactuca sativa* var. Parris Island) was used in even-numbered cycles. Supplemental N was added to columns (as  $\text{Ca}(\text{NO}_3)_2$  solution to Cycle 5 and as  $\text{NH}_4\text{NO}_3$  solution in Cycle 6) during Cycles 1–6 to maintain target total available N levels of 80–120 kg/ha for romaine and 40 kg/ha for oats. K was added (as KCl solution) at 80 kg/ha for all but natural pH control columns in Cycle 6. Crops were harvested at 11–14 weeks after seeding, representing the ‘green chop’ harvest stage for oats and clover, and maturity for romaine.

Columns were watered weekly during cropping cycles by filling the reservoirs previously described. Percolate was collected and sampled 2–3 days after watering, by which time all percolation had ceased. During any extended idle periods between cropping cycles, columns were covered with aluminum foil, and limited amounts of deionized water (up to 0.5 l/week) were applied to columns as needed to keep columns from desiccating. However, additions were limited so that percolate would not be produced between cycles. Supplemental lighting was used to extend day lengths by 4–8 h during fall and winter months, but was in general minimized to prevent excessive evaporation/transpiration rates. The greenhouse was lightly whitewashed in summer to help control temperatures and reduce ventilation requirements. Additional circulation fans were used to minimize temperature variations within the greenhouse.

### 2.5. Analytical

Soil samples (collected as described above) were air-dried at 55°C. Fine roots and other plant matter were removed, and the samples were ground in a porcelain mortar and pestle, sieved through a 16-mesh plastic screen to remove any coarse fragments (all soils were largely free of stones and pebbles), and stored in polyethylene bags. Soil pH was determined in 1:1 soil/distilled water suspensions, mixed at 0 and 0.5 h and measured at 1 h. Reference electrode errors were reduced by placing the reference electrode in the supernatant above the settled soil suspension during measurement.

Percolate was collected weekly during operating cycles. Percolate volumes are expressed as depth (cm) of percolate (volume divided by the surface area of the soil columns). Total percolate mass was determined by weighing collection jugs in the greenhouse, and 125-ml subsamples were taken. Electrical conductivity (EC) and pH analysis was typically carried out either immediately, or within 24 h, and 35-ml subsamples were frozen. Mass-weighted monthly composite samples for metals

analysis were produced from these frozen subsamples. During Cycles 6–8, the monthly composite samples were again proportionally composited to form a single sample for each column that represented percolate from the entire cropping cycle. Samples were agitated during collection and were vortex-mixed at each stage of the compositing process. Samples were filtered through coarse acid-washed cellulose filters (Fisher Scientific Q8, 10  $\mu\text{m}$  porosity), and filtrates were analyzed for metals and other elements via inductively coupled argon plasma (ICP) spectroscopy using a Thermo-Jarrell-Ash Model 975 ICP unit at Cornell University’s Nutrient Analysis Laboratory. All results are expressed as the mean and standard deviation of the triplicate columns for each treatment.

At the end of Cycle 5 the percolate collection jugs were rinsed with 30 ml of 4 M HCl to test for potential metal deposition in the jugs. Rinsates were digested at 80°C for 16 h. A representative subsampling of 10 columns with detectable percolate metals losses as of Cycle 5 were analyzed via ICP spectroscopy after filtration with coarse acid-washed cellulose filters. The mass of metals recovered were compared with cumulative percolate metals losses as of the end of Cycle 5. Similarly, the drainage tubing of four columns (old site Hudson, and Arkport soil dewatered sludge, NV, and natural control treatments) was replaced at the end of Cycle 7. The original tubing was scraped and acid-rinsed (4 M HCl) to remove a dark brown plaque-like coating. Rinsates were digested at 80°C for 16 h, filtered and analyzed via ICP spectroscopy. Metals recovered were compared with cumulative percolate metals losses as of the end of Cycle 8.

Statistical testing of the significance of observed effects was limited by the substantial interaction of independent variables (sludge treatments with soil pH). In view of this and the ongoing nature of the study, conclusions were limited to readily observable trends.

## 3. Results

This paper presents percolate results and soil pH levels observed during the first eight cropping cycles of this ongoing study. Primary comparisons are among sludge products, soil types and initial pH levels. Comparisons are also made between old site Hudson soil and Hudson control soils.

### 3.1. Percolation rates

Percolation rates—expressed as mean weekly depth (cm/week)—varied markedly over the course of each cropping cycle, decreasing steadily and, in many cases, ceasing as transpiration increased as a result of crop growth. Following harvest, percolation would resume

once soil moisture levels recovered. Mean weekly percolate depth (cm/wk) for the Arkport soils (Fig. 2) were typically greater than for the Hudson soils during active crop growth. This was a result of the finer Hudson soil's higher water-holding capacity, which enabled the Hudson soil columns to retain and store a larger fraction of applied water, reducing percolate volumes. Arkport columns with significant crop yields often began exhibiting signs of water stress at the end of each weekly watering cycle, whereas this rarely occurred with Hudson soils. Treatments with lower crop yields (particularly controls) tended to have correspondingly greater percolate masses. Variation in percolation rates between cropping cycles was the result of a number of factors,

including crop, temperatures of greenhouse and ventilation air, humidity and amount of supplemental lighting, all of which affected the rate of transpiration and thus percolation. In most cases, percolation rates were 50–150% of the target of 30 cm per cycle, equivalent to the mean annual recharge rate in New York State. Old site Hudson column percolation rates (Fig. 3) tended to be greater than comparable controls due to lower crop yields.

### 3.2. Percolate EC

EC values, used as an index of solution concentrations, were summarized as volume-weighted means for

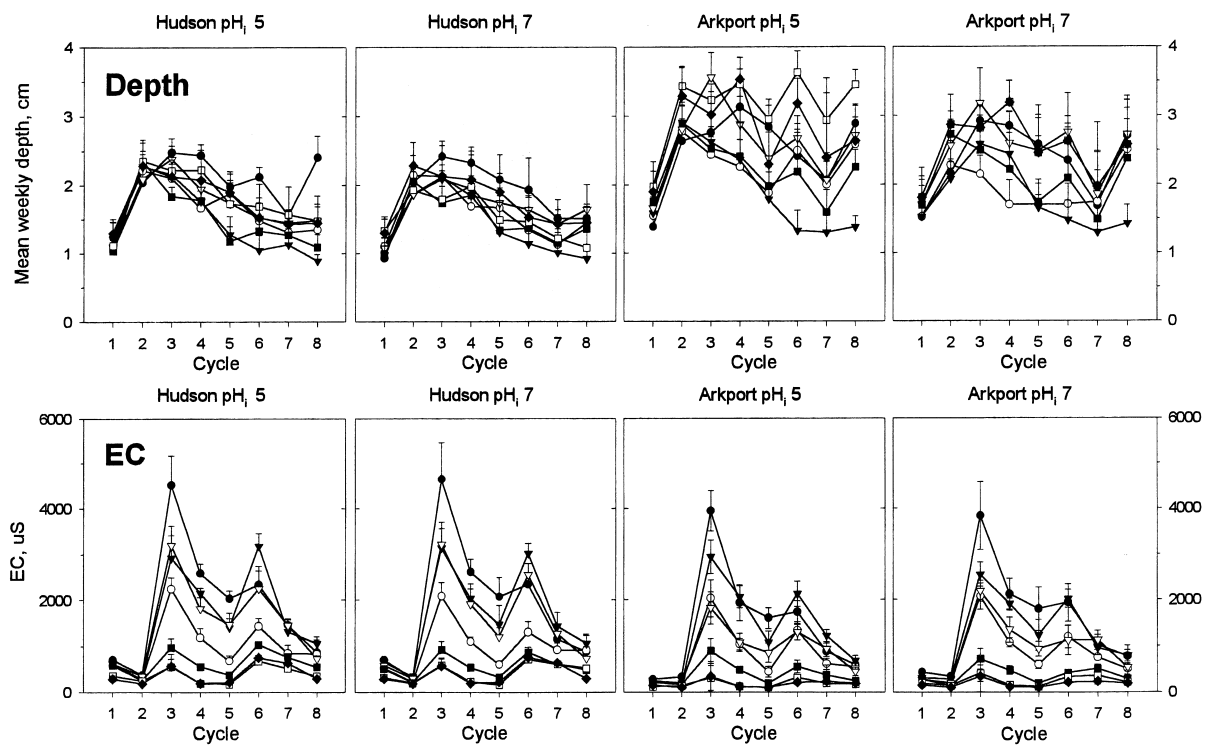


Fig. 2. Hudson and Arkport column percolate depth (cm) and electrical conductivity (EC) ( $\mu\text{S}/\text{cm}$ ), grouped by soil and initial soil pH. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

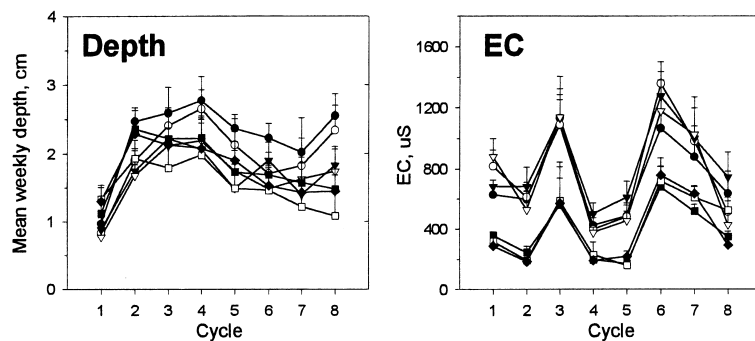


Fig. 3. Old site Hudson (OS) and Hudson control (H) column percolate depth (cm) and electrical conductivity (EC) ( $\mu\text{S}/\text{cm}$ ), plotted by soil and initial pH: ●, OS5; ○, OS7; ▼, OS natural; ▽, OS >7; ■, H5; □, H7; ◆, H natural.



each cropping cycle (Fig. 2). Dewatered sludge caused the largest increases in EC, followed closely by pellets, N-Viro and compost. Ash had relatively little effect on percolate EC. The peaks during Cycles 3 and 6 were not attributable to the nutrient solution additions made to the columns, since the natural control columns were given no nutrient supplementation and showed relative increases similar to the columns. The increases seem to be associated with the extended idle periods immediately preceding both cycles. Examination of weekly EC results (data not shown) show that levels were elevated at the beginning of these cycles, and steadily declined for all treatments. It is possible that the interim waterings that preceded each cycle translocated salts, making them available for rapid leaching once regular full waterings resumed. Old site column percolate EC varied markedly over time (Fig. 3), apparently due to the inter-cycle idle periods prior to Cycle 3 and 6 discussed above. Levels were greater than controls, but were well below levels observed in the newly sludge-applied columns.

### 3.3. Percolate pH

Percolate pH results for the Hudson and Arkport soils varied markedly with treatment and time (Fig. 4). For Hudson columns, heavy sludge loadings in Cycles 3 and 4 resulted in sharp decreases in percolate pH for

columns loaded with dewatered and pelletized sludges. This likely resulted from oxidation of loaded S and N (supported by percolate S data presented later), both of which are strongly acidifying reactions. Percolate pH levels were still recovering as of Cycle 8. Compost depressed percolate pH slightly, and ash had little effect. N-Viro resulted in delayed increases in percolate pH. Cycles 5–8 saw a slight downward trend in percolate pH for most treatments, possibly due to gradual effects of the acid rain application. Arkport soil, being more poorly buffered, saw steeper declines in percolate pH for dewatered and pelletized sludge, reaching levels as low as pH 4.0. Compost depressed pH more significantly than in the Hudson columns, and increases due to N-Viro did not occur until Cycle 7. There was no apparent effect of the pre-Cycle 8 lime additions to pH 7 columns except for slight increases in Cycle 8 percolate pH for the Arkport compost and control columns. Old site Hudson column (Fig. 5) percolate pH values generally remained in a narrow range from pH 6.0 to 6.5 despite differences in soil pH treatments.

### 3.4. Soil pH

Soil pH (Fig. 4) was determined on samples taken initially (prior to any adjustment in soil pH) and at the end of each cropping cycle. Dewatered sludge columns saw pH levels decline somewhat during agronomic

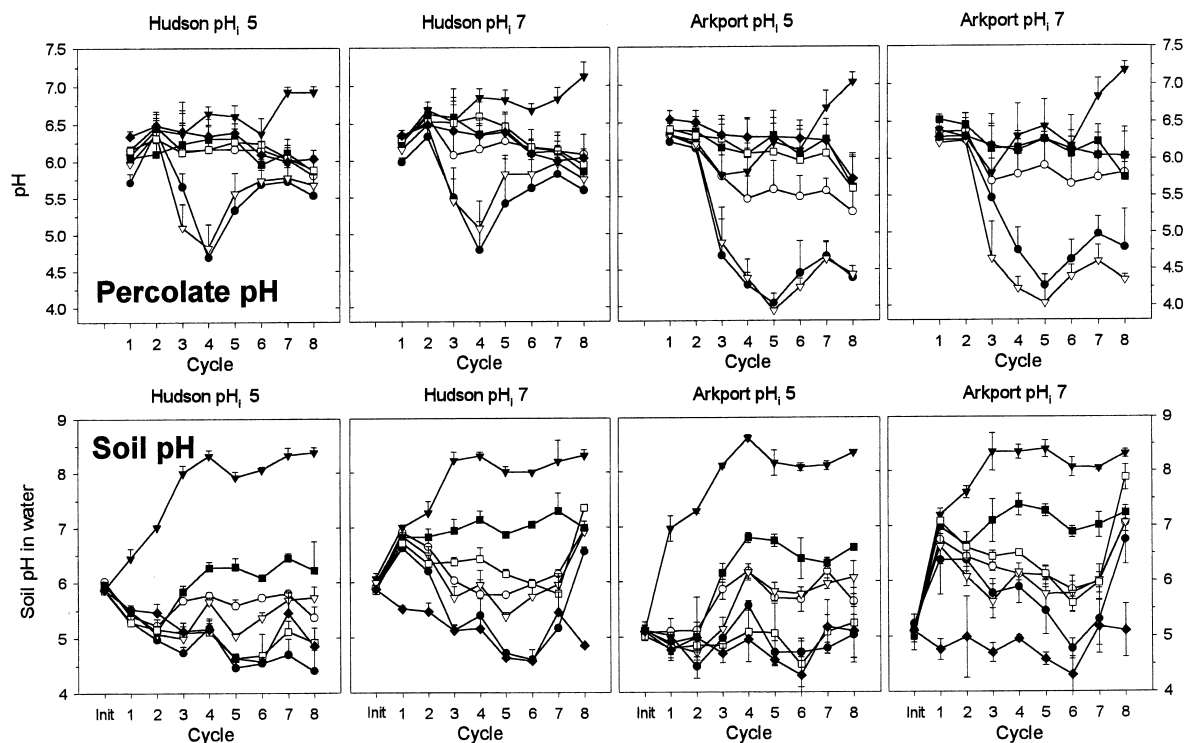


Fig. 4. Hudson and Arkport soil column percolate pH and soil pH, grouped by soil and initial soil pH. Sludge treatment: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

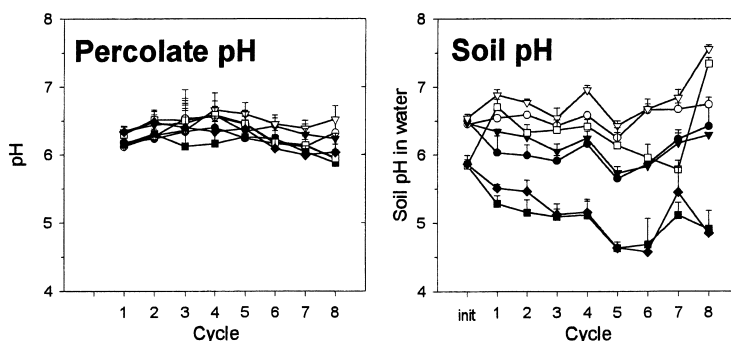


Fig. 5. Old site Hudson (OS) and Hudson control (H) soil column percolate and soil pH, plotted by soil type and initial soil pH: ●, OS5; ○, OS7; ▼, OS natural; ▽, OS > 7; ■, H5; □, H7; ◆, H natural.

sludge loadings (Cycles 1 and 2), followed by substantial declines resulting from the heavy loadings of Cycles 3 and 4. The decline continued through Cycle 6. The depression in pH was again attributed to N and S oxidation. The pH levels of 4.5–4.8 as of the end of Cycle 6 may have been buffered against further declines by the organic matter present. Compost applications had a much less dramatic effect on pH levels, with low pH columns actually increasing to over pH 5.5 by Cycle 6. High pH columns declined to 5.6–6.0. Increases in Cycle 8 in pH 7 columns were due to lime reapplications.

Pellets had pH trends similar to compost. It should be noted that at the end of Cycle 8 many pellets were still largely intact: soil-coated but firm and black-colored inside, which may explain why soil pH effects were not more similar to those of dewatered sludge. N-Viro raised all soil pH levels to 7 by the end of Cycle 2 and over pH 8 by Cycle 3. Slight differences among soil pH treatments remained until Cycle 5, but by the end of Cycle 8 all treatments were between pH 8.0 and 8.3, which is approximately the maximum pH that a carbonate-dominated system in equilibrium with atmospheric CO<sub>2</sub> can sustain. Ash exerted an alkaline effect on soils, although less dramatic than N-Viro. Control columns showed a steady decline throughout the study as a result of the synthetic acid rainfall. By the end of Cycle 7, the Hudson and Arkport pH 7 controls had nearly returned to their pre-adjustment levels, indicating that the initial lime additions had nearly been consumed. Hudson natural and low pH controls declined to 4.6–4.7 by the end of Cycle 6, with Arkport natural and low pH controls slightly lower. The slight increases seen in Cycle 7 levels may have been linked to overall lower percolate volumes during the cycle.

Old site soil columns (Fig. 5) showed substantial buffering capacity in their resistance to acid or lime additions during pH adjustment, with the low pH treatment rebounding to pH 6 in Cycle 1 and the high pH treatment reaching only pH 6.8. Over the course of the cropping cycles the high pH and pH 7 treatments converged at circa pH 6.7 while the natural control and low pH treatments converged at pH 5.8.

### 3.5. Percolate metals

The initial leaching (carried out prior to any pH adjustment or sludge application) resulted in little or no detectable metals in Hudson or Arkport soil percolates (Table 5). Percolate metal concentrations (volume-weighted means) for the entire Cycles 1–8 sequence are summarized in Table 6. Time-series plots of mean percolate concentrations of most analytes are presented in Figs. 6–11. Graphs have similar y-axis scaling to facilitate comparisons among soil treatments.

Percolate concentrations of Cu (Fig. 6) were greatest for N-Viro treatments, mirroring the pattern (although not the magnitude) of short-term mobilities observed in TCLP testing of sludge. (N-Viro TCLP mobilities were 50, 43 and 24% of total metals for Mo, Cu and Ni, respectively.) Concentrations peaked between 0.3 and 0.65 mg/l following the heavy loadings of Cycles 3 and 4, decreasing below 0.1 mg/l by Cycle 8. As discussed elsewhere (Richards et al., 1997), this is likely due to transport of Cu–organic complexes mobilized by organic matter dissolution resulting from elevated pH. All other sludge treatments had peak concentrations below 0.05 mg/l, and overall mean concentrations below 0.025 mg/l.

Table 5  
Initial baseline leaching ICP analysis results, mean values (as mg/l) for each group of soil columns

Element	Hudson	Arkport
Ag	nd <sup>a</sup>	nd
Cd	nd	nd
Cr	nd	nd
Cu	nd	nd
Mo	nd	nd
Ni	nd	nd
P	1.31	1.10
Pb	nd	nd
Zn	0.005	0.001

<sup>a</sup> nd, Not detected.

Table 6  
Hudson (H) and Arkport (A) soil column mean percolate concentrations for Cycles 1–8 (mg/l)<sup>a</sup>

Sludge	pHi		B		Ca		Cd		Cr		Cu		K		Mg		Mn	
			H	A	H	A	H	A	H	A	H	A	H	A	H	A		
DW	5	m	0.161	0.081	362	248	0.001	0.004	0.025	0.021	0.010	0.013	31.8	8.6	56.2	35.9	2.13	6.24
		sd	0.007	0.011	9	12	0.000	0.001	0.002	0.002	0.002	0.001	6.8	1.5	3.8	3.2	0.41	2.05
	7	m	0.178	0.079	353	285	0.001	0.002	0.025	0.021	0.012	0.011	28.4	7.8	50.3	34.4	1.52	3.46
		sd	0.050	0.009	15	37	0.001	0.000	0.001	0.003	0.001	0.002	10.1	2.3	5.3	3.7	0.70	0.77
COM	5	m	0.232	0.079	163	128	0.001	0.001	0.018	0.015	0.018	0.019	19.5	5.3	33.1	21.9	0.05	0.15
		sd	0.025	0.011	2	20	0.000	0.000	0.001	0.002	0.002	0.002	5.7	1.0	2.1	4.3	0.02	0.04
	7	m	0.184	0.092	162	149	0.001	0.001	0.016	0.015	0.016	0.017	19.5	7.0	26.7	19.1	0.08	0.15
		sd	0.022	0.008	11	16	0.000	0.000	0.000	0.000	0.003	0.003	7.2	2.9	2.1	0.0	0.07	0.00
NV	5	m	0.290	0.242	278	223	0.001	0.002	0.023	0.021	0.112	0.120	40.7	9.4	49.5	37.5	0.16	1.15
		sd	0.016	0.004	17	25	0.001	0.000	0.002	0.003	0.030	0.015	14.5	1.2	4.2	1.9	0.04	0.47
	7	m	0.285	0.255	303	256	0.001	0.001	0.023	0.020	0.081	0.151	34.8	9.3	55.3	33.0	0.05	0.84
		sd	0.051	0.005	34	12	0.000	0.000	0.004	0.002	0.015	0.021	18.2	2.2	10.3	0.5	0.01	0.40
PELL	5	m	0.197	0.064	276	150	0.001	0.003	0.022	0.016	0.017	0.024	31.3	10.3	41.1	20.0	1.28	3.86
		sd	0.032	0.005	33	31	0.000	0.001	0.001	0.002	0.001	0.003	5.0	2.0	1.4	5.3	0.34	0.73
	7	m	0.230	0.066	295	181	0.001	0.002	0.020	0.014	0.018	0.019	33.1	8.2	38.4	17.2	0.78	3.41
		sd	0.032	0.006	11	29	0.000	0.001	0.002	0.004	0.004	0.003	22.1	1.8	7.1	3.5	0.28	0.88
ASH	5	m	0.239	0.099	110	65	0.001	0.001	0.013	0.008	0.011	0.008	18.0	3.2	23.5	10.0	0.07	0.06
		sd	0.029	0.008	8	12	0.000	0.000	0.001	0.002	0.001	0.002	3.9	0.7	1.2	1.1	0.05	0.01
	7	m	0.217	0.103	90	65	0.001	0.001	0.010	0.007	0.009	0.006	21.5	6.7	16.0	8.3	0.02	0.03
		sd	0.019	0.009	4	5	0.000	0.000	0.001	0.001	0.001	0.001	10.5	4.4	0.9	0.7	0.00	0.00
CTRL	5	m	0.148	0.037	56	26	0.001	0.001	0.008	0.005	0.009	0.008	14.5	2.2	10.9	3.9	0.03	0.07
		sd	0.032	0.006	6	3	0.000	0.000	0.002	0.000	0.001	0.002	2.8	1.0	1.5	0.2	0.01	0.07
	7	m	0.178	0.063	54	37	0.001	0.001	0.008	0.006	0.012	0.010	19.6	3.0	10.1	4.8	0.02	0.11
		sd	0.061	0.011	13	4	0.000	0.000	0.001	0.001	0.002	0.003	4.5	1.3	3.5	0.3	0.00	0.11
	Nat	m	0.179	0.051	48	24	0.001	0.001	0.007	0.006	0.010	0.008	18.7	1.7	9.4	3.2	0.02	0.03
		sd	0.033	0.006	4	2	0.000	0.000	0.001	0.001	0.001	0.001	6.1	0.3	0.9	0.1	0.00	0.00
			Mo		Na		Ni		P		Pb		S		Zn			
			H	A	H	A	H	A	H	A	H	A	H	A	H	A		
DW	5	m	0.001	0.002	7.92	6.00	0.009	0.095	0.765	0.360	0.001	0.004	115.7	65.1	0.108	0.204		
		sd	0.000	0.000	0.39	0.30	0.002	0.029	0.102	0.030	0.001	0.002	4.4	2.5	0.026	0.096		
	7	m	0.002	0.002	7.66	6.24	0.011	0.054	0.557	0.365	0.002	0.003	105.9	80.8	0.068	0.060		
		sd	0.001	0.000	0.86	0.09	0.002	0.006	0.095	0.027	0.001	0.000	5.7	13.4	0.007	0.025		
COM	5	m	0.002	0.002	6.25	4.43	0.006	0.025	0.828	0.299	0.003	0.005	86.5	52.2	0.011	0.019		
		sd	0.000	0.000	0.23	0.36	0.002	0.006	0.213	0.046	0.001	0.002	2.9	5.5	0.001	0.006		
	7	m	0.003	0.002	6.01	4.65	0.006	0.035	0.577	0.248	0.002	0.004	81.4	55.8	0.010	0.023		
		sd	0.001	0.000	0.25	0.40	0.002	0.010	0.219	0.048	0.000	0.000	5.0	3.0	0.003	0.007		
NV	5	m	0.012	0.009	10.22	8.01	0.022	0.062	0.532	0.382	0.002	0.005	118.2	113.3	0.014	0.012		
		sd	0.003	0.005	0.24	0.66	0.010	0.015	0.058	0.086	0.001	0.001	18.7	16.1	0.006	0.004		
	7	m	0.011	0.013	10.88	8.32	0.015	0.059	0.578	0.363	0.002	0.005	117.2	109.2	0.011	0.017		
		sd	0.001	0.005	1.35	0.14	0.001	0.021	0.124	0.025	0.001	0.001	7.9	9.5	0.003	0.006		
PELL	5	m	0.002	0.003	7.08	4.78	0.009	0.068	1.100	0.370	0.002	0.006	108.4	64.1	0.051	0.053		
		sd	0.001	0.001	0.59	0.51	0.002	0.009	0.222	0.065	0.000	0.003	17.4	15.6	0.006	0.018		
	7	m	0.002	0.002	7.16	4.77	0.009	0.079	0.882	0.342	0.001	0.004	102.8	70.0	0.031	0.056		
		sd	0.000	0.000	1.36	0.60	0.003	0.029	0.241	0.056	0.000	0.001	15.2	4.7	0.010	0.040		
ASH	5	m	0.003	0.003	5.07	2.33	0.005	0.007	0.547	0.538	0.003	0.006	64.9	35.5	0.012	0.010		
		sd	0.001	0.001	0.23	0.02	0.001	0.002	0.053	0.369	0.000	0.001	10.5	3.0	0.003	0.002		
	7	m	0.007	0.002	4.30	2.39	0.003	0.004	0.457	0.243	0.003	0.005	55.3	36.2	0.007	0.007		
		sd	0.002	0.001	0.18	0.19	0.000	0.000	0.073	0.056	0.001	0.001	1.7	4.7	0.000	0.001		

(Table 6 continued on next page)

Table 6 (continued)

			Mo		Na		Ni		P		Pb		S		Zn	
			H	A	H	A	H	A	H	A	H	A	H	A	H	A
CTRL	5	m	0.002	0.002	2.90	1.47	0.004	0.005	0.521	0.326	0.003	0.007	20.5	5.1	0.009	0.007
		sd	0.001	0.000	0.61	0.06	0.001	0.002	0.145	0.057	0.000	0.002	3.4	1.4	0.002	0.001
	7	m	0.002	0.002	3.29	2.03	0.005	0.010	0.948	0.240	0.004	0.007	9.9	9.9	0.009	0.008
		sd	0.000	0.001	0.11	0.36	0.001	0.002	0.602	0.037	0.000	0.001	2.0	4.0	0.001	0.002
Nat	m	0.002	0.002	3.14	1.66	0.004	0.006	0.781	0.258	0.003	0.008	10.4	5.1	0.009	0.009	
	sd	0.001	0.000	0.23	0.09	0.000	0.001	0.157	0.075	0.001	0.001	1.0	0.3	0.001	0.002	

<sup>a</sup> DW, dewatered digested sludge; COM, composted sludge; NV, alkaline-stabilized sludge; PELL, dried sludge pellets; ASH, incinerated sludge ash; CTRL, control; m, mean; sd, standard deviation.

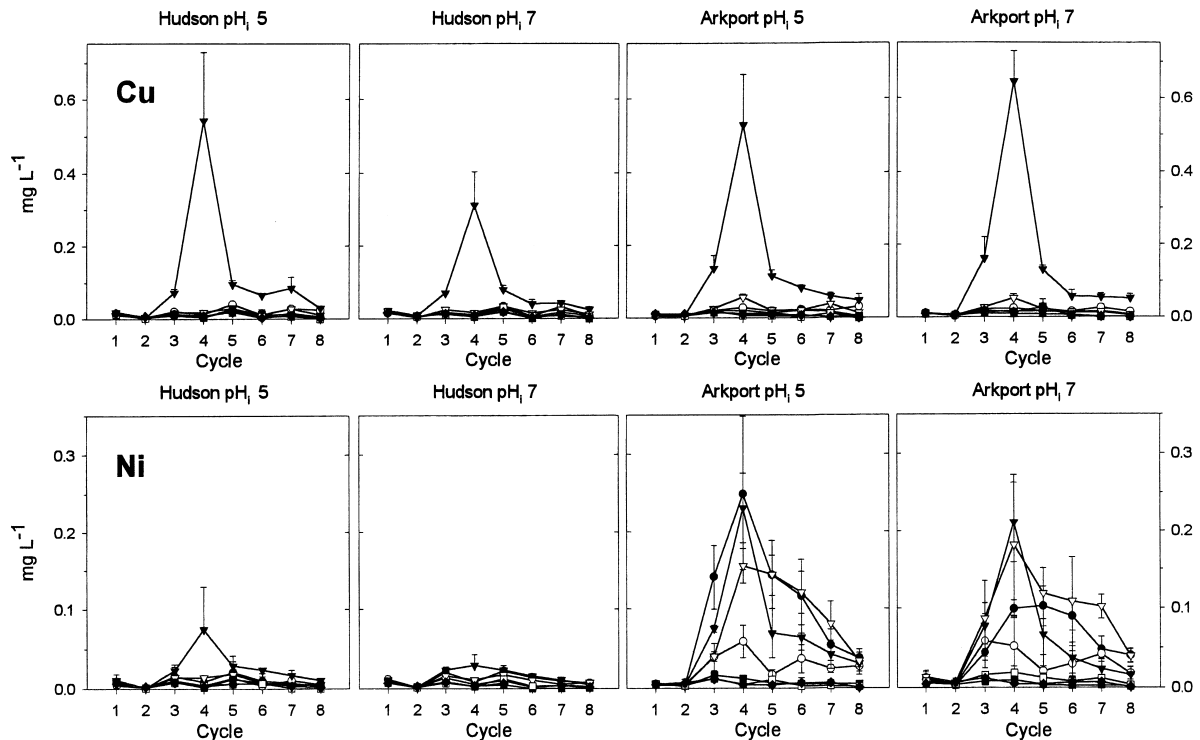


Fig. 6. Hudson and Arkport soil column percolate Cu and Ni. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

Ni percolate concentrations (Fig. 6) varied strongly with soil type. For Hudson soils, the only notable Ni mobility came from N-Viro (again mirroring TCLP results) during Cycle 4, with greater concentrations observed from the pH 5 columns. Arkport soils had markedly greater concentrations beginning with Cycle 3. In the low pH columns, dewatered sludge, pellets and N-Viro had the greatest concentrations, peaking in Cycle 4. Compost treatment percolates peaked in Cycle 4 at levels far lower than the other treatments but equivalent to the greatest concentrations observed for Hudson soils. For pH 7 columns, dewatered sludge concentrations were lower while peak N-Viro and pelletized sludge percolate concentrations were similar to pH 5 levels. Compost followed a pattern similar to the pH 5 columns.

Cd concentrations (Fig. 7) were near lower detection limits for all Hudson soils, but showed increases in percolates from Arkport soils applied with dewatered and pelletized sludge products during heavy loadings in Cycles 3 and 4. Zn varied greatly with soil type and pH. In Hudson soils, dewatered sludge had the greatest percolate concentrations, reaching 0.24 mg/l in Cycle 4 for the low pH soil, and decreasing somewhat in Cycle 5. Pelletized sludge Zn concentrations were approximately one half of dewatered sludge levels. However, concentrations from the dewatered sludge treatments reached 0.7 mg/l in low pH Arkport columns, although those from pelletized sludge were similar to Hudson soil results. Levels in the pH 7 columns were also similar to the Hudson pH 7 columns.

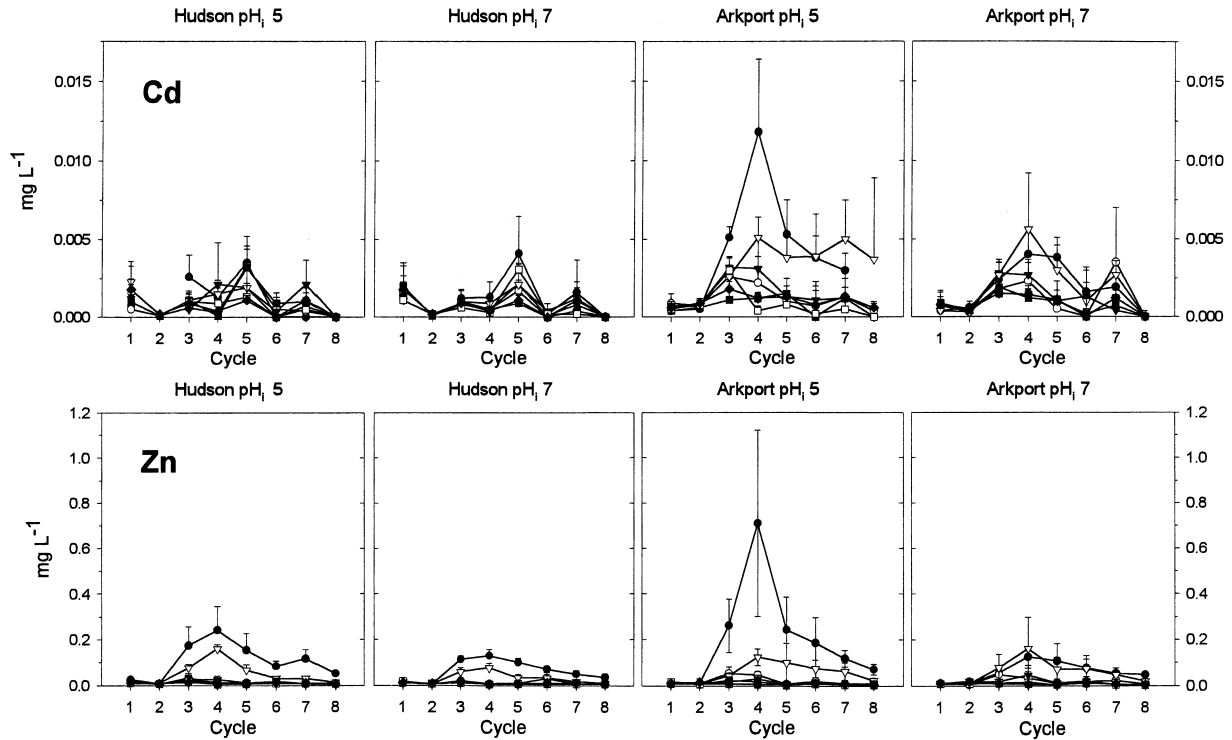


Fig. 7. Hudson and Arkport soil column percolate Cd and Zn. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

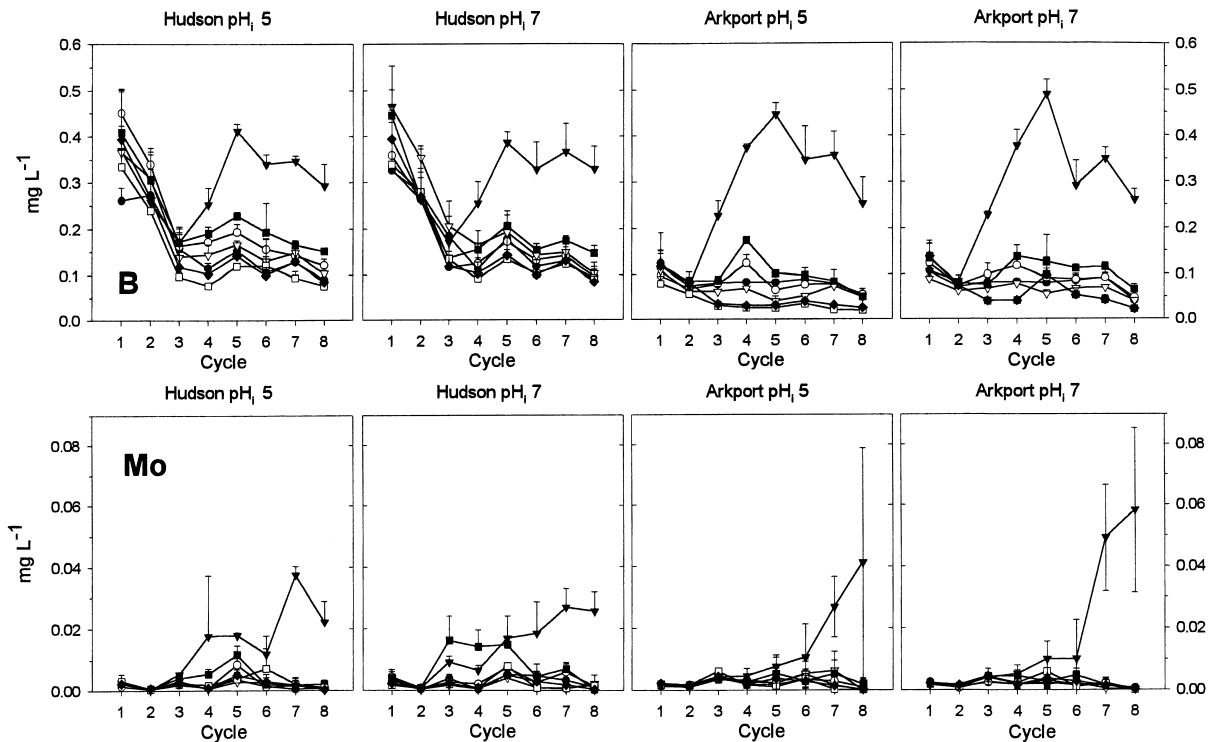


Fig. 8. Hudson and Arkport soil column percolate B and Mo. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

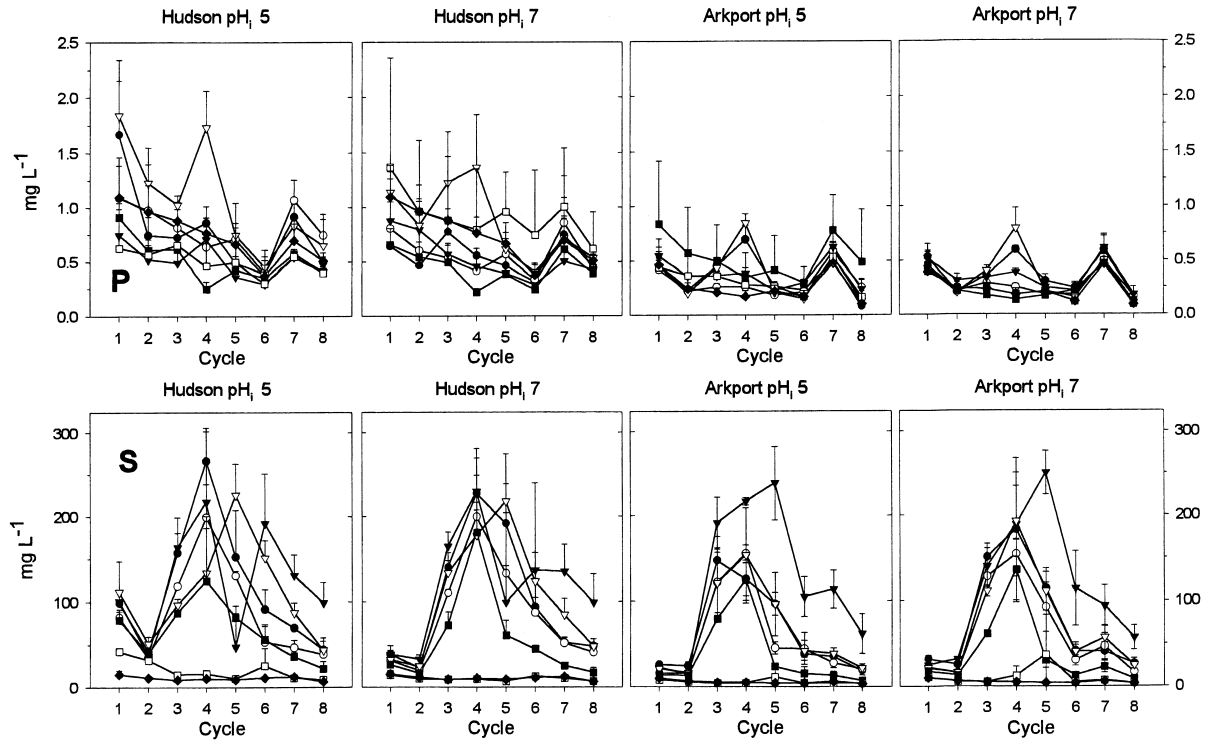


Fig. 9. Hudson and Arkport soil column percolate P and S. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

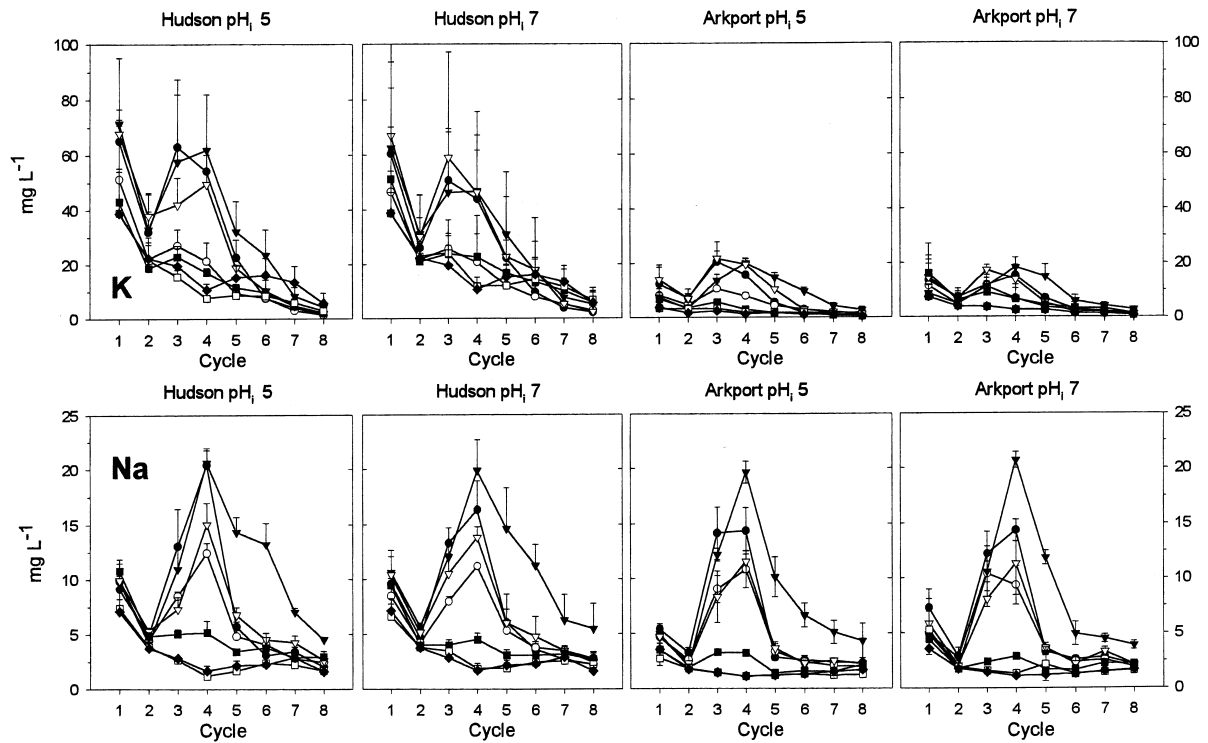


Fig. 10. Hudson and Arkport soil column percolate K and Na. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

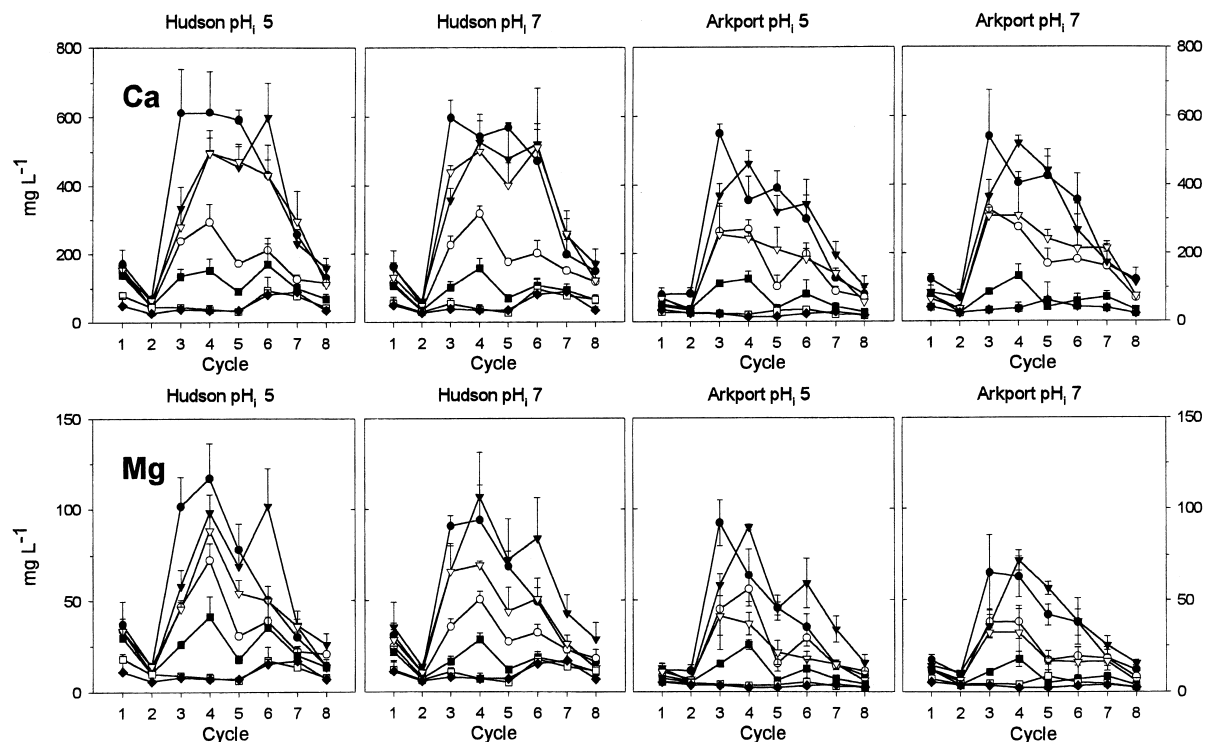


Fig. 11. Hudson and Arkport soil column percolate Ca and Mg. Sludge treatments: ●, dewatered digested sludge (DW); ○, composted sludge (COM); ▼, alkaline-stabilized sludge (NV); ▽, dried sludge pellets (PEL); ■, incinerated sludge ash (ASH); □, control; ◆, natural control.

B concentrations (Fig. 8) were greatest during initial leachings, possibly due to aerial deposition of B from the University coal-fired heating plant, which was upwind of the fields from which the soils were extracted. Concentrations decreased steadily, stabilizing by Cycle 4 for all treatments but N-Viro. B concentrations from N-Viro columns increased steadily from Cycles 3 to 5, reaching 0.5 mg/l in Arkport soils, slightly higher than Hudson soils. Mo concentrations increased from Cycle 3 for all N-Viro treatments, reaching 0.03 mg/l for Hudson soil treatments. Increases in Arkport soils were delayed until Cycle 6, when levels began increasing steeply, with large variability among triplicates. Ash also had elevated Mo concentrations in Hudson soil columns, particularly in the pH 7 columns.

P concentrations (Fig. 9) declined over the course of operation. The greatest concentrations observed during the heavy loadings were from pelletized sludge on Hudson soils, but concentrations were generally indistinguishable from control levels. Percolate S were elevated in pH 5 Hudson soils due to acid additions during pH adjustments (not needed by pH 5 Arkport columns due to low initial pH levels). Concentrations from all sludge treatments increased during the Cycle 3 and 4 heavy loadings, exceeding 100 mg/l. All levels declined subsequently, with levels persisting in Arkport soil N-Viro columns.

K concentrations (Fig. 10) declined steadily for all control columns, although initial concentrations were

greatest in Hudson columns. During Cycles 3 and 4, K concentrations increased for all sludge products. Increases in Hudson column percolates were greatest for dewatered sludge, N-Viro and pellets (peaking between 40 and 60 mg/l) but were small for compost and ash. Levels were lower in Arkport soils but concentrations followed the same relative pattern. High initial levels of Na mirrored results seen with B, declining steadily in all control columns, again suggesting a uniform deposition source while soils were still in the field. Concentrations stabilized in all controls during Cycles 4 and 5. Slight increases in percolate Na were observed from ash additions for both Hudson and Arkport soils. Concentrations in dewatered, pellets, compost and N-Viro treatments peaked during Cycle 4, and declined in Cycle 5, although N-Viro levels declined more slowly.

Ca percolate concentrations (Fig. 11) followed patterns that, for a given soil type, were similar for both pH levels. For Hudson soils, concentrations increased from dewatered sludge columns to approximately 600 mg/l for Cycles 3–5. Concentrations were nearly as great from pellets and N-Viro. High Ca concentrations from the N-Viro percolates are due to the substantial Ca loadings. However, in the case of dewatered sludge and pellets, it is unknown how much of the Ca leached originated from the sludge itself and how much was mobilized from the soil due to the strong acidification that took place as a result of heavy loadings, as evidenced by percolate and soil pH levels. Compost additions resulted

in increases to 300 mg/l percolate Ca by Cycle 4. In Arkport soils, Ca concentrations from dewatered sludge peaked in Cycle 3. N-Viro results were similar to that seen with Hudson soils, but pelletized sludge additions resulted in percolate Ca concentrations that were substantially lower, similar to composted sludge. Ash results were similar to Hudson soils. Baseline control levels in the pH5 and natural control treatments continued to decline, falling below 25 mg/l. Mg concentrations followed a pattern similar to Ca, although concentrations were about one-fifth of those seen with Ca.

Percolate Cr concentrations were low at approximately 0.02 mg/l for all sludge-treated columns except ash, with levels of 0.01 mg/l for ASH and control columns (Table 6). Pb concentrations were near lower instrumental detection limits. Mn concentrations were greatest for soil columns treated with dewatered and pelletized sludges, followed by N-Viro treatments. Concentrations were greater from Hudson soils than from Arkport soils.

Percolate concentrations for the old site Hudson columns (Figs. 12 and 13) showed no clear time-related trends for most elements. B, K and Na did follow the trend observed in the Hudson and Arkport control column percolates, indicating leaching of apparently aerially deposited materials. Despite apparent trends toward lower Cu, Ni and Zn concentrations in the highest soil pH treatment, analysis of variance performed on mean results from Cycles 1 to 5 (data not

shown) found that the old site soil pH treatments had no significant effect ( $p=0.05$ ) on percolate concentrations. The only exception was S, which was elevated in the low pH treatment simply due to sulfuric acid additions during pH adjustment. When all old site columns were considered as a single treatment, mean concentrations of all analytes were significantly different from the Hudson control percolates during Cycles 1–5, as determined by analysis of variance ( $p=0.05$ ). Unlike all other analytes, P concentrations were greater in the control percolates.

For all treatments tested, metals recovered by the acid-rinsing of the percolate collection jugs were less than 1% of the cumulative percolate losses except for P and Pb. P recoveries ranged up to 1.4% of cumulative losses for Hudson soil columns. Pb recoveries—which were low—nevertheless represented up to 3.5% of percolate losses for Hudson and Arkport columns, and up to 10% for old site Hudson treatments, possibly a result of transport of a small amount of lead arsenate-contaminated topsoil (from old orchard pesticide sprays) through the column.

The acid-washing and digestion of plaque lining the drain tubing of four columns yielded variable results. Recoveries for the dewatered sludge- and N-Viro-treated columns were less 2.2% of cumulative Cycles 1–8 percolate losses except for Cr recoveries, which were 4–4.8% of percolate losses. Mass recoveries from the Arkport natural control were similar. Old site column

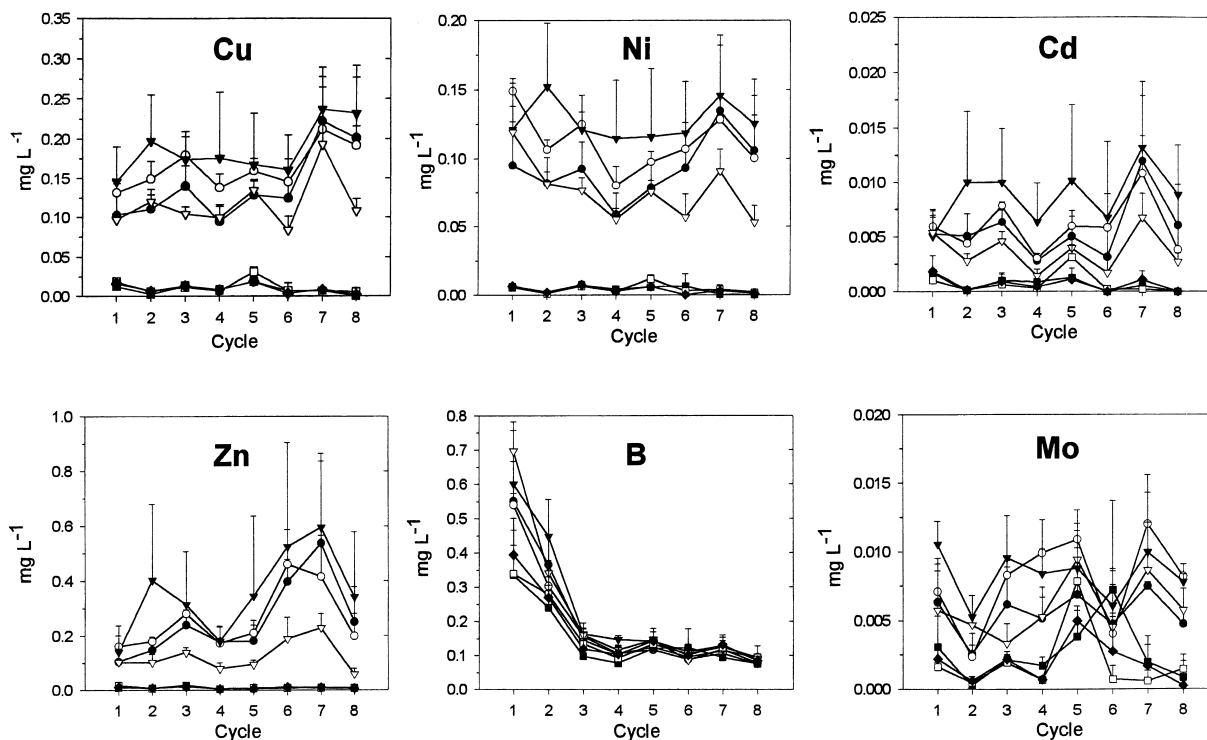


Fig. 12. Old site Hudson (OS) and Hudson control (H) soil column percolate Cu, Ni, Cd, Zn, B and Mo, plotted by soil and initial soil pH: ●, OS5; ○, OS7; ▼, OS natural; ▽, OS >7; ■, H5; □, H7; ◆, H natural.



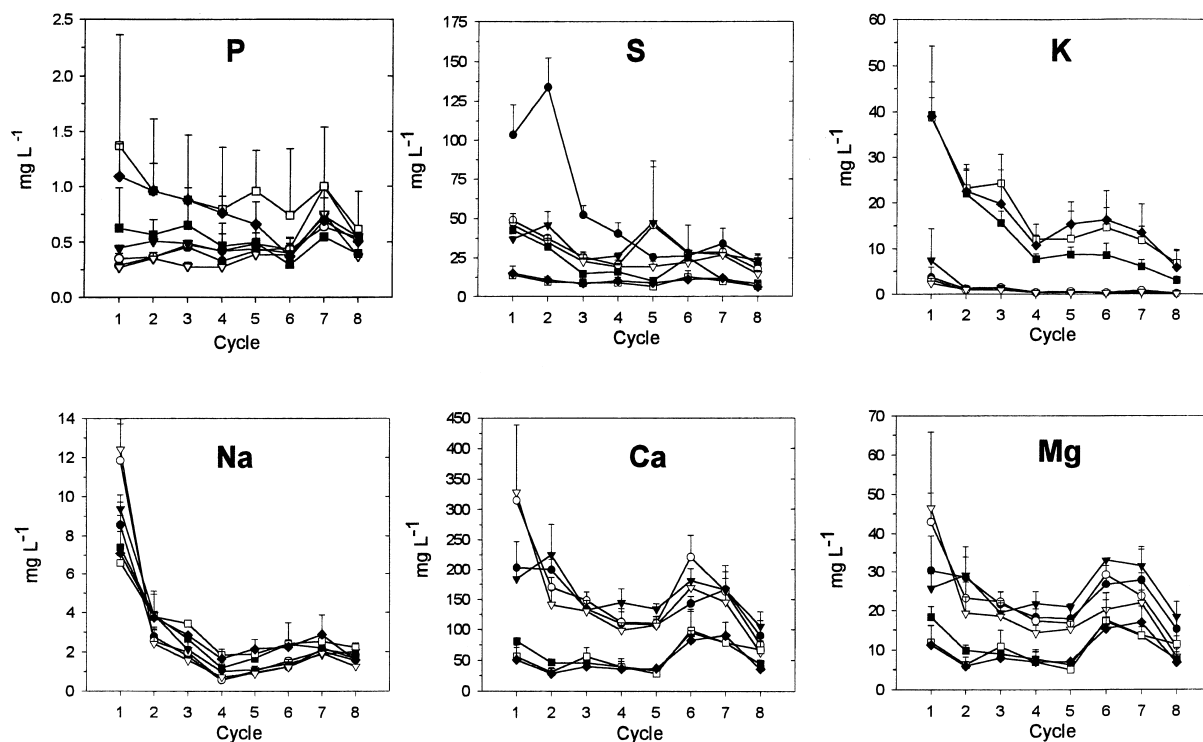


Fig. 13. Old site Hudson (OS) and Hudson control (H) soil column percolate P, S, K, Na, Ca and Mg, plotted by initial soil pH: ●, OS5; ○, OS7; ▼, OS natural; ▽, OS > 7; ■, H5; □, H7; ◆, H natural.

recoveries were below 2% of percolate losses except for a 9.3% Pb recovery.

#### 4. Discussion

The sludge application experiment demonstrated that the mode of sludge processing, soil type and initial soil pH strongly affected metal mobility. The sludge loading rates used were, by design, much heavier than agronomic use would dictate over the time frame of the experiment. The heavy loadings were used to build soil metal contents to levels corresponding to approximately 30 years of agronomic applications. Cumulative dry matter loading rates for dewatered sludge, pellets and compost were, also, similar in magnitude to rates used for land reclamation (Sopper, 1993). However, due to the low metal contents of the sludge products used, metals loading rates were substantially lower than recommended loading limits associated with land reclamation. The loading rates used in this study (using the greatest rate among sludge products in Table 3) ranged from 9 (Cr, Pb) to 74% (Zn) of the recommended maximum loading rates for reclamation of land to be used as farmland reported in Sopper (1993). Loading rates were also substantially below the cumulative Part 503 loading limits (USEPA, 1993), ranging from 1 to 10% of limits for all elements shown in Table 7 but Mo. Loadings were 39% of the previous Mo limit (subse-

quently withdrawn by USEPA). Dry matter loadings were approximately one-third of the cumulative loadings used by Dowdy et al. (1991), and on the same order of magnitude as those used by Chang et al. (1984).

The use of normalization factors (Table 3) ensured that elemental loading rates were comparable among sludge products, only differing significantly when processing resulted in large elemental additions (Ca, Mg and K in additives for N-Viro stabilization) or selective losses (such as Cd volatilization during incineration). The equal relative loading rates do not, however, represent equivalent agronomic time frames. The loadings of dewatered sludge, pellets and compost simulated approximately 30 years of agronomic applications. In contrast, the 663 tons/ha cumulative loading of N-Viro, intended for use as a lime substitute, is probably closer to several hundred years of field applications (which are texture- and initial pH-specific), as reflected in the elevated soil pH levels observed. (Logan et al., 1997a, observed a similar soil pH after 500 tons/ha loadings). Ash would not normally be used for land application, and would in this case be prohibited due to Mo concentrations exceeding Part 503 ceiling limits (Richards et al. 1997).

Aside from the expected effects of heavy N-Viro applications, the extreme depression of pH by dewatered sludge was more pronounced than anticipated. With a high Ca sludge loaded at rates similar to this study, Logan et al. (1997b) observed only a one unit

Table 7

Comparison of elemental loading rates with recommended maximum reclamation loading rates (for farming use) and USEPA Part 503 cumulative loading limits

Element	Farmland reclamation limit (Sopper, 1993) (kg/ha)	Part 503 maximum cumulative limit (USEPA, 1993) (kg/ha)	Maximum cumulative loading rate used in this study:		
			from Table 3, (kg/ha)	as % of reclamation limit	as % of Part 503 limit
Cd	3.4	39	1.4	41	3.6
Cr	336	3000	30	9	1.0
Cu	840	1500	134	16	8.9
Mo	–	[18] <sup>a</sup>	7.1	–	39
Ni	33	420	8.5	26	2.0
Pb	336	300	30	9	10
Zn	168	2800	125	74	4.5

<sup>a</sup> Mo limit subsequently withdrawn.

decrease in soil pH in a well-buffered soil over the course of several years, followed by a gradual recovery. However, Harrison et al. (1996) reported depression of soil pH to 4.5 following a 300 tons/ha sludge loading.

Sludge-processing mode, soil type, soil texture and time since application had substantial effects on percolate metal mobilities following heavy sludge loadings. Metals and nutrients had a range of response patterns, indicating that observed mobilities were not simply due to washing of sludge products through the soil columns. Analytes had unique patterns of response to one or more factors in the sludge/soil/soil pH treatment matrix. B, Cu and Ni leaching was most prominent from N-Viro and was relatively insensitive to soil pH or type. Mo leaching was greatest from N-Viro, followed by ash for Hudson soils. Ni percolate concentrations were greatest from dewatered sludge and pellets applied to low pH Arkport soils, followed by pH 7 soils and N-Viro treatments. Zn showed sensitivity to all three treatment variables, with greatest concentrations from dewatered sludge and pellets; among these sludge treatments, Arkport soils had greater concentrations than Hudson, and for each soil type the lower initial pH had greater concentrations. Ca trends were similar to those of Mg, with greatest leaching from dewatered sludge, N-Viro and pellets. Relatively little leaching of P was observed from any treatment, similar to the results of Jackson et al. (1999). The observed mobilities of Cu, Ni and, in more recent cycles, Mo mirrored TCLP results (Richards et al., 1997), although on a diminished scale.

Present results indicate that composted sludge and ash had the lowest overall trace metal mobilities. Pelletized sludge results were similar to the dewatered sludge, with notable leaching of Ni, Cd and Zn. N-Viro had the widest range of leachable elements, including Cu, Ni, B and Mo. Peak Cu, Ni and Mo levels from N-Viro

exceeded the old site columns concentrations, as did B concentrations (once old site percolate concentrations stabilized). This pattern is similar to the results of Theis et al. (1998) who used the same sludge products under more intensive leaching conditions.

The overall cumulative percolate losses to date have been, at most, a small percentage of total applied metals. It should be remembered that percolate concentrations reported here reflect only soluble metals (free or complexed to soluble organics), since coarse filtration was required prior to ICP analysis. Tests of several column systems indicated little adsorption of HCl-soluble metals in drain tubing or collection jugs, but the potential for deposition in the bottom of the soil column system cannot yet be assessed. The extent of mixing of sludge with the surface soil layer may have been greater in this study than would occur with plowing in the field. The lack of the presence of earthworms (which were not added to columns due to potential for percolate contamination and increased management requirements) is also a difference from field conditions: worms could be expected to open or maintain flow paths as well as process several of sludge products.

The old site column experiment did not demonstrate significant soil pH effects on metal mobility due to wide variation among replicate columns. As cited in the Experimental approach section, columns with visible variations in soil (marbling or veins of residual sludge) were intentionally distributed among the various pH treatments. This resulted in variable initial soil metal concentrations among soil columns. Initial topsoil metal analysis (data not shown) resulted in coefficients of variation (among soil columns) of 10–12% for Cd, Cu and Zn.

Perhaps the most significant finding with the old site columns was the agreement of observed percolate concentrations with lysimeter results measured at the field

Table 8

Percolate metals concentrations (mg/l) greenhouse old site soil columns (native pH, mean of Cycles 1–5 and 1–8) in comparison to field site sludge plot lysimeter samples

Element	Old site soil columns				Old site sludge plot lysimeter samples	
	Cycles 1–5		Cycles 1–8		Mean	SD
	Mean	SD	Mean	SD		
Ca	164	14	160	12	177	43
Cd	0.008	0.005	0.009	0.004	0.008	0.000
Cr	0.029	0.003	0.032	0.003	0.033	0.008
Cu	0.176	0.055	0.188	0.052	0.132	0.013
Ni	0.127	0.038	0.128	0.036	0.199	0.008
P	0.470	0.070	0.505	0.048	0.398	0.158
Zn	0.279	0.178	0.351	0.209	0.444	0.034

site. Table 8 compares mean percolate concentrations of several key elements from the natural pH soil columns (which are identical to the soil at the field site) to mean old site data accumulated over several years (Richards et al., 1998). Greenhouse soil column data is presented for two time frames (cumulative means of Cycles 1–5 and of Cycles 1–8) to demonstrate that the agreement is not coincidental. Results are nearly identical to the field site, with the greatest difference being Zn, with mean percolate concentrations of 0.28–0.35 mg/l from the soil columns versus 0.44 mg/l in the field site. As was the case with the field site (Richards et al., 1998), the present percolate fluxes represent a small fraction of soil total metals, so it can be expected that these percolate concentrations could be maintained long term.

Several additional years of operation are planned for the experimental system (with no additional sludge additions). As was evident from recent Mo and B trends, the system is still in flux. Soils in pH 7 treatments were restored to near-initial pH levels by lime additions prior to Cycle 8, while low pH treatments will be allowed to acidify, simulating unmanaged conditions. Future work will include determination of mobile forms of metals (free, colloid-adsorbed or organically complexed), and soil mass balances. Implications of metal mobility for long-term groundwater quality also need to be examined.

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