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Effects of Sludge Application on the Cd, Pb and Zn Levels of Selected Vegetable Plants

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Tissue of Swiss chard (*Beta vulgaris* L.), radish (*Raphonus sativus* L.) and tomato (*Clycopersicon esculentum* Mil.) were examined to determine the effects of repeated sludge applications on the accumulation of Cd and Zn. As many as six crops of Swiss chard and radish and three crops of tomato were cultured in greenhouses during the 15-month experiment. Results showed that the plants did not extract appreciable Cd or Zn from the soil when the soil metals were at indigenous levels. Following the initiation of sludge applications, elevated levels of Cd and Zn were detected in all harvested plant tissues. For Swiss chard and radish, the leaf Cd and Zn concentrations increased with the amount of applied sludge. In tomato, elevated Cd and Zn levels were observed following the initial sludge application; however, subsequent applications did not increase the levels significantly. When crops were planted in soils no longer receiving sludge application, plant tissue continued to exhibit Cd and Zn levels greater than non-sludged controls in all six croppings. If the soil received only a single large dosage of sludges at the beginning and then was continuously cropped for the remainder of the experiment, the highest Cd and Zn content of the plant tissue occurred at the first crop harvest. After the first cropping, the metal concentration in the plant tissue of successive crops was reduced to a stable, but still elevated level (a level greater than the non-sludged controls). There was no indication that the elevated plant-tissue Cd and Zn contents caused by the sludge applications would be reduced to the background level after the termination of sludge disposal.

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INTRODUCTION

IN THE UNITED STATES, cropland application of sewage sludges—primarily in small communities—has been practiced since the advent of wastewater treatment. More recently, wastewater treatment authorities in many densely populated areas have also become interested in adopting land-oriented approaches to sludge disposal. Despite its growing popularity, waste disposal on land has seldom been adequately regulated and monitored. Concerns have been expressed repeatedly over the possible release of potentially hazardous biological agents and chemical constituents into the environment through such disposal. Besides warning about potential public health hazards associated with pathogens, scientists have expressed cautions about the fate and effects of sludge-bound heavy metal elements in soils (Page, 1974; Chaney et al., 1976).

Other studies have shown that introducing heavy metals into the soil through sludge disposal often accelerates their uptake by plants. Under certain soil chemical conditions, elevated heavy metal levels in soil resulted in serious plant injuries (Vandecaveye, Horne, and Keaton, 1936; Soane and Saunder, 1959; Lee and Page, 1967). Even before the plant itself exhibits any phytotoxic symptoms, however, plant tissue accumulation of heavy metals might reach levels considered hazardous to consumers (CAST, 1976, 1980). Among the commonly occurring sludge-borne heavy metal elements, cadmium (Cd) and Zinc (Zn) were the most likely to accumulate in plant tissue.

Field and greenhouse experiments have demonstrated that Cd and Zn in plant tissue always increased with incremental rates of metal-laden sludges. When sludge is applied repeatedly at a given site, the common cause and effect relationship (input of heavy metal elevates levels of such metals in plant tissue) did not necessarily hold. In such case, the growing crops were no longer influenced by a simple, one-shot sludge treatment; rather, they were affected simultaneously by both the most immediate sludge input and the previous deposits as well. Thus, it would be impossible to distinguish the contribution of each deposit on the heavy-metal content of the harvested crops.

Recently, several long-term studies suggested that the Cd and Zn content of crops harvested from repeatedly sludged soils tended to be a function of the most immediate sludge input, rather than of the cumulative total metal content of the soil (Bates et al., 1975; Dowdy et al., 1978). Other studies showed that the elevated heavy metal levels of affected plant tissue consistently declined with each successive crop growing cycle following the termination of sludge disposal (Hinesly et al., 1977; Hinesly et al., 1979). Baker et al. (1979) and Chaney (1976) demonstrated continuously high levels of plant tissue metal content once the soil became contaminated.

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These data indicate that heavy metals deposited in the soil under certain conditions exhibit diminishing effects on the growing plants. If this diminishing effect could be confirmed experimentally, it would have a major influence on formulating a strategy to minimize the hazard potentials of heavy metal elements during land application of sludges. Assuming (1) a gradually diminishing impact on the growing crops and (2) the ability to maintain the chemical condition of sludge-treated soil properly, growers could apply sludge on a given soil for an extended time period without increasing the heavy-metal hazards to growing plants. If the potentially hazardous effects in such soils were cumulative, the soil's capability to receive metal-laden sewage sludge would be seriously restricted. Since heavy metals were not very mobile in the soil, they would affect plant growth for many years. The present study was designed to use several plants as indicators to examine residual heavy-metal effects in sludge-affected soils using several plants as indicators.

EXPERIMENTAL PROCEDURES AND METHODS

Plants from three vegetable crops were grown in a glasshouse for 15 months on sludge-amended soils in 11-liter ABS plastic potting containers. The composted sludge was obtained from Joint Water Pollution Control Plant (JWPCP) of County Sanitation Districts of Los Angeles County. Physical and chemical properties of the sludge are given in Table 1.

Four levels of sludge treatments representing various residual effects were used:

(a) no sludge (or other metal-containing substance) added to soils;
(b) repeated sludge application: soil received composted sludge equivalent to 45 mT/ha before each cropping;
(c) residual sludge effects—soils were recovered from experimental plots where sludges had been applied. Before the experiment, each soil received a total of 135 mT/ha of JWPCP composted sludge over a 24-month period in three equally spaced applications. No additional sludge was added during the glasshouse experiment. This treatment represented residual effects of heavy metals following termination of sludge disposal;
(d) single sludge application: soils received a single massive dosage of sludge equivalent to 135 mT/ha. The soil was then cropped repeatedly without further sludge addition.

Two types of soils were used: (a) Domino loam (fine-loamy, mixed, thermic Xerollic Calciorthid), calcareous soil with 20% clay; (b) Greenfield sandy loam (coarse-loamy, mixed, thermic Typic Haploxeralf), non-calcareous soil with approximately 10% clay.

Properties of these soils are summarized in Table 2. Three species of vegetable plants were used: (a) Tomato (*Lycopersicon esculentum* Mil.); (b) Swiss chard (*Beta vulgaris* L.); (c) Radish (*Raphanus sativus* L.).

Under the same sludge treatment, these plants accumulated different amounts of metals in plant tissue. For details of the effects on various plant species, see Bingham et al. (1975).

All treatment combinations were replicated four times. After soils were treated with respective amounts of sludge, 12 kg of sludge-soil mixture was added to each potting container, and moisture was equilibrated. Radish was seeded directly in the pot and thinned to 10 plants after germination. Swiss chard and tomato were germinated first in seed germination cubes and transplanted later, so that pots contained either four Swiss chard or one tomato. During the growing season, each pot was watered according to tensiometer readings to maintain the soil moisture regime between 20 to 80 centibar soil moisture tension. After each growing cycle, crops and soils were sampled, and the soil in each pot was allowed to dry out gradually. Soil from four replications of the same treatment was combined, crushed, mixed, treated with sludge (if needed), repotted, and planted. Throughout the growing period the glasshouse temperature was maintained between 20 °C to 30 °C. Six crops of radish and Swiss chard and three crops of tomato were grown during the 15-month experimental period. Crops were harvested when they reached maturity (tomato) or reached a predefined size (radish and chard). No attempt was made to compare the crop yield of each treatment.

Atomic adsorption spectrophotometric techniques were used to determine heavy metals (Cd, Zn, and lead (Pb)) in plant tissue and soils. Soil metals were determined by taking 4 grams of soil sample that had been refluxed overnight in 10 ml of 4 N HNO_3 at 70 °, diluting it to volume and filtering it. For plant tissue analysis, 200-mg samples were digested by a HNO_3-$HClO_4$ mixture (Ganje and Page, 1974).

To make our task more manageable we decided to make a composite of plant tissue from four replicates of the same treatment combination on the equal weight basis. Before consolidating any sample, we made a preliminary analysis to compare the composite and the individual replicates of each data set. Comparison showed that the values

Table 1. PHYSICAL AND CHEMICAL CHARACTERISTICS OF COMPOSTED SLUDGES

Constituent	Concentration
Total solids	65%
Fixed solids (% total solids)	72%
Total nitrogen	0.93%
Total phosphorus	1.39%
Cadmium	61 μ g/gm
Chromium	1,856 μ g/gm
Copper	1,042 μ g/gm
Nickel	482 μ g/gm
Lead	1,997 μ g/gm
Zinc	3,547 μ g/gm

Table 2. PROPERTIES OF THE EXPERIMENTAL SOIL

Item	Domino loam	Greenfield sandy loam
pH	7.8	6.5
Organic Matter (%)	4.3	0.8
Cation Exchange Capacity (meq/100 gm)	20	6.5
Cadmium* (μ g/gm)	0.95	0.8
Chromium* (μ g/gm)	15.5	14.0
Copper* (μ g/gm)	21.0	14.5
Nickel* (μ g/gm)	13.5	11.0
Lead* (μ g/gm)	14.5	12.0
Zinc* (μ g/gm)	75.0	64.5
Soil texture (%)		
clay	22	10
silt	37	27
sand	41	63

*4 M HNO_3 extractable.

from analyzing the composite samples were closely correlated with the mean of the four replications. Correlation coefficients ranged from 0.965 to 0.998. On the basis of these results, it was felt that the loss of information due to sample composition would be minimum. Statistically, data were analyzed using the analysis of variance with disproportionate subclass numbers (Ostle, 1963).

RESULTS AND DISCUSSION

Theoretically, two approaches may be used to assess the impact of heavy metals on crops grown in sludge-affected soils. The potential for plant tissue to accumulate excessive amounts of heavy metals may be estimated by determining the contents of plant-available heavy metals in the soil. Because the concentration of heavy metal elements usually is low, their chemical activities and, therefore, plant availability must be influenced by other more abundant soil chemicals. For this reason, the total soil metal content is hardly an indicator of plant availability. We know that pH, cation exchange capacity, organic matter content and iron oxides of the soil all affect the heavy metal concentrations of the plant tissue; however, the exact role of each soil chemical constituents on plant availability cannot be properly identified.

Many empirical procedures may be used to diagnose the deficiency and/or toxicity of trace metal elements in cropland soils; nevertheless, none can predict the magnitude of plant tissue accumulation of heavy metals. Therefore, to project the plant tissue accumulation of heavy metals indirectly by measuring their concentrations in the soil has not been entirely successful. A more direct approach is to measure the accumulation of these metals in plant tissue harvested from affected soils. When that is done, the heavy metal content in plant tissue reflects the chemical interactions of the soil throughout the entire growing season. This approach to the measurement has also been difficult. Under the same treatment, plant species appeared to respond differently. Even for the same species, there may be seasonal variation. More important, this approach completely ignored the chemical mechanisms that make metals available for absorption. Thus, results should not be extrapolated beyond the range where observations were taken. All these constraints have made it virtually impossible to quantitatively predict the plant tissue accumulation of heavy metal elements in sludge-affected cropland soils.

Data presented in Tables 3 through 10 summarize Cd, Pb and Zn contents of both the sludge-affected soils and the harvested plant tissue at the end of each growing cycle. Soil pH associated with each cropping cycle was also tabulated. The pH of the Greenfield sandy loam and Domino loam varied from 5.6 to 6.9 and 6.9 to 7.3, respectively. Soil pH of less than 6.5 occurred only in the Greenfield sandy loam soil that was not treated with sludges. Large amounts of Pb were also added into the soil through sludge applications. Although Pb content of harvested plant tissue varied from 0.50 μ g/gm to 7.3 μ g/gm, lead level in the plant tissue did not appear to be affected by sludge inputs. For soils not contaminated by sludge applications, the mean Pb concentration of plant tissue and its standard deviation were 2.25 μ g/gm and 1.05 μ g/gm, respectively. In the sludge-affected soils, plant tissue Pb content averaged 2.56 μ g/gm and the standard deviation was 1.29. For this reason, the sludge-borne lead and its effects on the plant tissue lead content were excluded from further discussion.

The effects of sludge applications on Cd and Zn content of harvested plant tissue were analyzed according to how the soil sludge was applied and they were examined through successive croppings. Since soil metal levels varied with sludge treatment, no attempt was made to compare the effects between treatments.

Crops grown on soils not affected by sludges

All natural soils contain trace amounts of heavy metal elements. The Cd content of agricultural soils in the U.S. rarely exceeds 1 μ g/gm; however, Zn content may vary

Table 3. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM A GREENFIELD SANDY LOAM SOIL NOT CONTAMINATED WITH SLUDGES

Plant	Cropping	pH*	Cadmium			Zinc			Lead		
			Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit
Swiss chard	1	6.6	0.60	0.30	—	63	48	—	12	1.5	—
	2	6.2	0.80	0.60	—	80	44	—	13	3.0	—
	3	5.6	0.50	0.70	—	60	40	—	10	1.0	—
	4	5.7	0.50	0.20	—	65	36	—	10	1.5	—
	5	5.6	0.60	0.30	—	68	114	—	12	3.9	—
	6	5.8	0.60	0.60	—	77	128	—	14	1.5	—
Radish	1	6.4	0.40	0.30	0.10	75	55	50	15	1.9	2.8
	2	6.3	0.80	0.80	0.10	75	79	74	20	2.1	1.8
	3	6.4	0.80	0.30	0.10	74	65	52	20	2.4	2.2
	4	6.3	0.80	0.30	0.10	70	75	45	20	1.8	0.5
	5	6.3	0.60	0.20	0.10	71	73	46	13	2.2	0.8
	6	6.4	0.60	0.50	0.10	81	73	79	12	1.2	1.0
Tomato	1	6.6	0.60	1.10	0.10	64	60	30	15	2.1	2.5
	2	6.4	0.70	1.40	0.10	65	85	30	20	4.0	3.5
	3	6.4	0.60	1.00	0.10	73	48	30	10	1.6	4.8

*pH of saturated soil paste measured at the end of each cropping.

Table 4. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM A DOMINO LOAM SOIL NOT CONTAMINATED WITH SLUDGES

Plant	Cropping	pH	Cadmium			Zinc			Lead		
			Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit
Swiss chard	1	7.2	0.8	0.10	—	88	70	—	18	2.9	—
	2	7.1	1.0	0.20	—	89	60	—	20	1.0	—
	3	7.0	1.0	0.20	—	90	50	—	20	1.3	—
	4	6.9	1.0	0.10	—	91	31	—	18	4.6	—
	5	5.7	0.6	0.10	—	90	71	—	15	3.2	—
	6	6.8	0.8	0.20	—	97	74	—	18	2.6	—
Radish	1	7.2	0.9	0.23	0.07	88	70	58	18	2.3	3.0
	2	7.2	1.0	0.14	0.08	89	88	50	30	1.8	1.3
	3	7.1	1.0	0.21	0.04	90	63	50	20	1.5	1.5
	4	7.1	1.0	0.20	0.10	91	63	35	25	1.5	1.0
	5	7.2	0.8	0.20	0.10	90	67	46	16	3.1	1.5
	6	7.3	0.8	0.23	0.12	97	62	47	19	1.5	1.4
Tomato	1	7.2	0.7	1.40	0.10	88	51	25	20	3.3	4.1
	2	7.2	1.0	0.90	0.10	93	73	29	20	2.5	2.3
	3	7.2	0.6	1.60	0.10	97	52	21	20	4.0	2.4

considerably, with concentrations ranging from 20 μ g/gm to 100 μ g/gm (Page, 1974). These levels of Cd and Zn did not induce any excessive accumulation of Cd and Zn in plant tissue (Page et al., 1980), whether crops were field- or greenhouse-grown. Each group of data points in Fig. 1 represents one complete cropping cycle. The lines on the graphs are not regression lines, but merely represent the mean Cd levels in soils and harvested plant tissues. They do not link data points; rather, they indicate the normal values and trend expected when the soils have not been affected by sludge applications or other means of Cd input. Although the two soils used for the experiment were different in both textural classification and Cd and Zn contents, there was little statistically significant difference in Cd and Zn levels in plant tissue between crops grown in either soil.

Crops grown on soils affected by sludge applications

Comparing the plant tissue Cd and Zn contents of the control treatment with those from sludge-treated soils showed clearly that the Cd and Zn concentrations of all plant tissue harvested from the sludge-affected soils were significantly elevated. The pattern

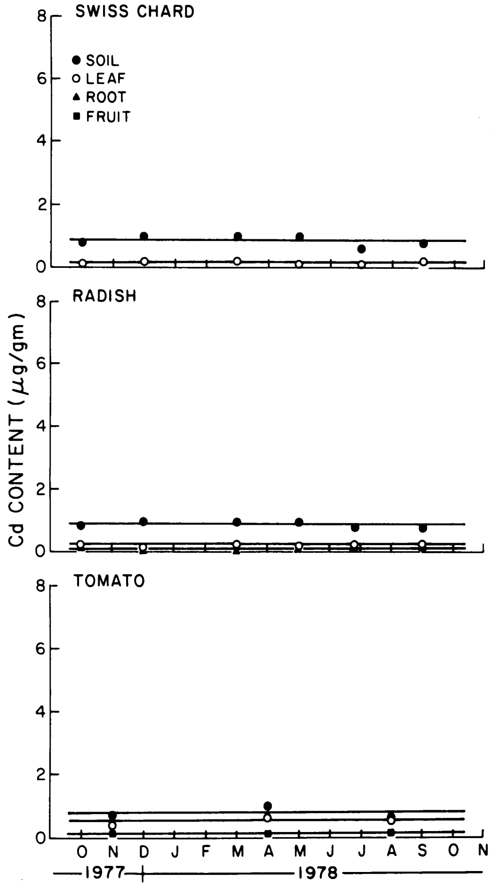

Fig. 1. Cd uptake of vegetable crops grown on the soil (Domino silty clay) not contaminated with heavy latent sludges.

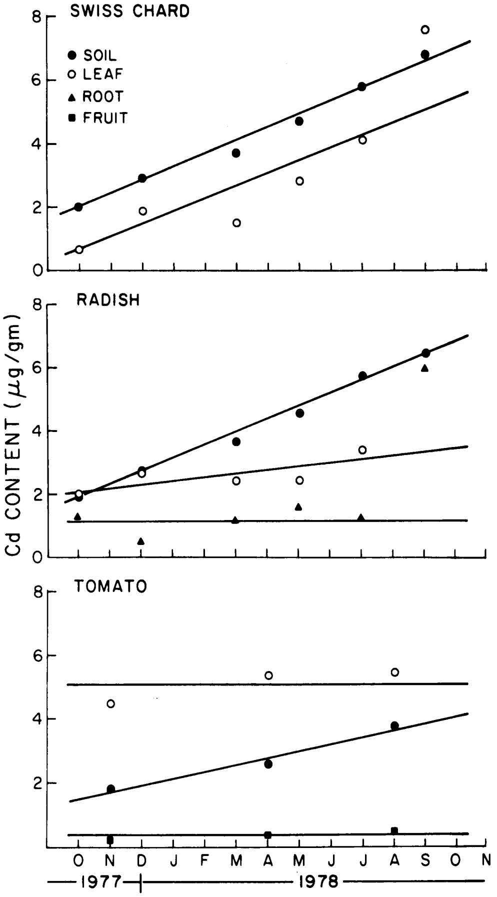

Fig. 2. Plant tissue Cd of vegetable crops grown in a repeatedly sludged soil (Domino silty clay).

of accumulating Cd and Zn into the plant tissue, however, appeared to vary with the nature of sludge treatments. This experiment was designed not to compare differences directly among sludge treatments or among crops, but to examine the plant tissue accumulation of Cd and Zn among successive croppings of the same sludge treatment. Statistical analysis indicated significant difference (at the 5% level) in plant tissue Cd and Zn contents between croppings when the soil was treated repeatedly with sludges as as well as when soil was treated with a single initial massive dose of sludge. How the plant tissue concentrations of Cd and Zn were affected by the continuous cropping will be discussed in the following sections.

Repeated sludge applications

In this treatment, sludges equivalent to 45 MT/ha were added into the soil before each cropping. At this application rate, amounts equivalent to 2.75 Kg/ha of Cd and 160 Kg/ha of Zn were incorporated into the soil with each cropping. After six cropping cycles, Cd content rose from the indigenous level to 6.0 to 7.0 μ g/gm, and Zn rose to more than 500 μ g/gm (Tables 5 and 6). Although plant tissue concentrations of Cd and Zn were high, crop injuries were not visible. Statistical analysis (Duncan's Multiple Range Test) comparing the metal content of plant tissue between croppings are summarized in Tables 7 and 8.

During the experiment, additional amounts of Cd and Zn were introduced into the soil with each sludge application, and contents of corresponding heavy metals in harvested plant tissue also appeared to rise; but there were not always statistically significant differences (at the 5% level) between the levels in plant tissue. In fact, only after the fifth or sixth cropping was there any statistically significant difference in metal content. Crops also responded differently when they were grown in different soils with the same

Table 5. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM THE GREENFIELD SANDY LOAM SOIL REPEATEDLY TREATED WTIH SLUDGES (45 mT/HA BEFORE EACH CROPPING)

Plant	Cropping	pH*	Cadmium†			Zinc†			Lead		
			Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit
Swiss chard	1	6.5	1.3	5.8xy	—	154	238x	—	84	1.0	—
	2	6.5	2.0	4.0x	—	245	281x	—	120	1.4	—
	3	6.5	3.0	2.3x	—	340	238x	—	157	1.5	—
	4	6.5	3.8	3.7x	—	430	274x	—	187	6.3	—
	5	6.4	4.6	3.2x	—	520	802y	—	198	2.8	—
	6	6.5	5.0	8.6y	—	610	1275z	—	226	3.2	—
Radish	1	6.5	1.4	2.3x	1.0x	158	325x	163x	60	1.3	1.5
	2	6.5	2.3	2.4x	1.0x	230	238x	111x	100	3.9	2.8
	3	6.5	3.2	3.3x	1.3x	315	291x	169x	160	3.8	6.8
	4	6.6	4.1	2.3x	1.3x	373	355x	134x	173	1.5	1.8
	5	6.5	5.2	2.3x	1.4x	430	322x	122x	227	1.3	4.6
	6	6.6	6.2	4.4y	3.6y	513	275x	114x	252	1.3	1.9
Tomato	1	6.5	1.4	6.3x	0.3x	175	155x	38x	55	2.1	4.1
	2	6.5	2.3	5.5x	0.4x	220	263x	50x	80	1.8	2.5
	3	6.5	3.0	5.5x	0.3x	255	173x	24x	110	1.8	3.2

*pH of saturated soil paste measured at the end of each cropping.

†Same letter following the plant tissue Cd and Zn content of each plant species indicated the metal content between croppings were not significantly different at 0.05 significant level.

sludge treatment. For example, the Cd content of Swiss chard grown in Domino soil rose gradually from 0.7 µ g/gm at the first cropping to over 9 µ g/gm at the sixth cropping (Table 6). On the Greenfield soil, however, Cd was higher to start with and remained at that level for three to four croppings, rising again during the sixth cropping to 8.7 µ g/gm (Table 5). This pattern of Cd accumulation was observed in both Swiss chard and radish. Because only three tomato crops were raised during the experimental period, conclusions for this crop were difficult to reach.

We could theorize that the metal concentration in the plant tissue indicated the elemental plant availability in the soil where the crop was grown. We also believed that levels of plant-available metals in the soils would rise gradually with each sludge application until they reached a saturation level for the given soil condition. If the soil chemical condition was kept unchanged, the plant-available metal would remain at the saturation level even if the sludge applications continued. The soil's chemical condition could also be altered by sludge input, causing the plant-available metal to reach a new level. Obviously, saturation level would vary with the soils and the metal elements. Based on this assumption, the observed plant tissue accumulation of Cd and Zn might be explained in terms of the plant availability in the soil.

Initially, plant availability of Cd in the sludge-treated Domino soil was low; but it rose steadily with the application of sludges. This indicated a continuous change in the soil chemical conditions with each sludge input. After the sixth cropping, there was no sign that a saturated plant-available Cd level had been reached. In the Greenfield soil, Cd was considerably more available to plants initially, remaining at that level for four to five cropping cycles, despite the repeated sludge inputs. At the sixth sludge application, crops accumulated significantly higher amounts of Cd. Thus, it appears that a change occurred in the soil chemical condition due to repeated sludge additions. The Cd con-

Table 6. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (µ g/gm) HARVESTED FROM THE DOMINO LOAM SOIL REPEATEDLY TREATED WTIH SLUDGES (45 mT/HA BEFORE EACH CROPPING)

Plant	Cropping	pH*	Cadmium†			Zinc†			Lead		
			Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit
Swiss chard	1	6.9	2.0	0.7x	—	178	95x	—	82	2.5	—
	2	6.8	2.9	1.9x	—	240	125x	—	95	1.5	—
	3	6.8	3.7	1.5x	—	385	166x	—	150	1.3	—
	4	6.7	4.7	2.8xy	—	450	336x	—	185	5.2	—
	5	6.6	5.8	4.10y	—	511	417y	—	220	3.0	—
	6	6.6	6.8	5.6y	—	573	665y	—	270	1.5	—
Radish	1	7.0	1.9	2.0x	1.3x	176	138x	69x	96	1.8	2.8
	2	6.8	2.8	2.8x	0.5x	260	138x	87x	170	2.0	1.5
	3	6.8	3.7	2.5x	1.2x	350	248x	94x	240	2.8	3.0
	4	6.7	4.6	2.5x	1.6x	425	322x	120x	260	2.0	2.3
	5	6.7	5.8	3.2x	1.3x	461	347x	120x	248	6.6	2.9
	6	6.7	6.5	10.8y	5.9y	533	650y	350y	277	1.8	1.9
Tomato	1	7.0	1.8	4.5x	0.20x	265	113x	31x	20	3.3	4.1
	2	6.8	2.6	5.4x	0.40x	330	135x	49x	20	2.5	2.3
	3	6.8	3.8	5.5x	0.50x	331	107x	37x	20	1.8	3.5

*pH of saturated soil paste measured at the end of each cropping.

†Same letter following the plant tissue Cd and Zn content of each plant species indicated the metal content between croppings were not significantly different at 0.05 significant level.

centrations of crops grown in the repeatedly sludge Domino soil are plotted in Figure 2. Lines on the diagram again were drawn to indicate the trend of Cd accumulation in plant tissue. The trend with Zn accumulations was very similar to that with Cd.

It was apparent that the pattern of accumulating Cd and Zn into crops grown on repeatedly sludged soil could be qualitatively described. Actual magnitude of plant tissue depositions, however, was more difficult to assess.

Table 7. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM A GREENFIELD SANDY LOAM SOIL WHOSE SLUDGE APPLICATION (135 mT/HA) WAS TERMINATED

Plant	Cropping	pH*	Cadmium			Zinc			Lead		
			Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit
Swiss chard	1	6.8	2.0	3.0	—	185	250	—	80	2.0	—
	2	6.6	2.3	3.2	—	185	288	—	90	1.8	—
	3	6.7	2.8	3.0	—	205	250	—	100	2.5	—
	4	6.6	2.5	3.0	—	201	205	—	78	1.6	—
	5	6.6	2.2	4.4	—	189	800	—	90	0.8	—
	6	6.4	2.3	4.6	—	187	925	—	87	1.2	—
Radish	1	6.8	1.9	3.5	0.40	185	163	91	80	2.5	4.3
	2	6.9	2.1	5.0	0.50	180	163	74	80	1.4	2.5
	3	6.8	1.9	3.8	0.30	210	145	72	108	2.4	2.5
	4	6.8	2.5	1.7	0.40	205	145	60	108	1.5	1.3
	5	6.8	2.1	1.3	0.04	187	170	62	78	5.3	2.9
	6	6.9	2.2	2.3	0.40	193	165	59	88	1.2	1.5
Tomato	1	6.8	2.2	5.2	0.40	185	164	33	88	2.3	5.0
	2	6.8	3.0	4.8	0.30	195	200	30	80	2.5	1.8
	3	6.8	2.2	5.8	0.30	187	175	34	80	1.5	3.2

*pH of saturated soil paste measured at the end of each cropping.

Table 8. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM A DOMINO LOAM SOIL WHOSE SLUDGE APPLICATION (135 mT/HA) WAS TERMINATED

Plant	Cropping	pH*	Cadmium			Zinc			Lead		
			Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit	Soil	Leaf	Root/ fruit
Swiss chard	1	7.1	2.0	1.8	—	224	198	—	82	6.3	—
	2	7.1	2.2	1.4	—	225	156	—	90	3.1	—
	3	7.0	2.5	1.5	—	220	142	—	190	1.5	—
	4	7.1	2.0	1.4	—	205	173	—	80	7.4	—
	5	6.9	2.4	1.8	—	240	175	—	98	2.1	—
	6	7.0	2.2	2.2	—	247	343	—	90	1.2	—
Radish	1	7.2	2.2	1.7	1.2	225	200	90	89	0.5	2.4
	2	7.2	2.2	0.9	1.1	225	175	75	90	1.4	2.4
	3	7.2	2.5	1.5	0.7	220	206	81	100	2.4	2.0
	4	7.3	2.5	1.6	0.5	205	188	70	190	1.5	2.5
	5	7.3	2.2	1.4	0.3	240	173	71	91	5.1	1.4
	6	7.3	2.2	1.6	1.2	247	182	119	89	1.4	2.1
Tomato	1	7.2	2.3	3.8	0.30	225	94	28	85	1.9	2.5
	2	7.3	2.4	5.5	0.30	210	128	25	80	4.0	2.3
	3	7.3	2.5	5.1	0.30	236	59	26	80	3.0	3.1

*pH of saturated soil paste measured at the end of each cropping.

Residual effects

Land application of sludges leads to the accumulation of significant amounts of heavy metals in soils. Since heavy metals are relatively immobile in the soil, they would remain there to affect growing crops long after sludge disposal itself ceased. To assess the potential hazards of heavy metals accumulation in plant tissue, it was essential to address their uptake by plants during both the actual sludge applicaton and the period after its termination. Soils in this part of the experiment were collected from two experimental sludge disposal sites adjacent to where the soils were obtained for the remainder of the experiment. Before being used for the observation of the residual effects, the soil received 8.24 Kg/ha of Cd and 479 Kg/ha of Zn from three sludge applications spanned over a 24-month period. When the treated soils were cropped, the resulting Cd and Zn content of harvested plant tissue was always higher than the control (Tables 7 and 8). But the elevated Cd and Zn content varied from one cropping to the other, even though metal concentrations in harvested plant tissue were not statistically significant (at the 5% level). After as many as six consecutive croppings, the Cd and Zn contents of all three crops remained elevated, and their concentrations showed little sign of decline.

Figure 3 was plotted to illustrate the trend established by continuously cropping those soils which had history of sludge disposal. Once the Cd and Zn contents of the soil were altered by sludge applications, it was difficult to predict (1) whether plant-available metals would return to the levels characteristic of those which occurred before sludge applications, or (2) how long would it take to reach a stabilized level if sludge application was terminated. Again, soil chemistry should play the most important role in determining the outcome. It appeared that once sludge disposal was terminated, the soil's chemical condition would reach equilibrium rapidly. For the two soils used in this ex-

Table 9. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μg/gm) HARVESTED FROM A GREENFIELD SANDY LOAM SOIL TREATED WITH A SINGLE SLUDGE INPUT (135 mT/HA) BEFORE CROPPINGS

Plant	Cropping	pH*	Cadmium†			Zinc†			Lead		
			Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit	Soil	Leaf	Root/fruit
Swiss chard	1	6.6	3.0	3.1x	—	204	388x	—	116	1.5	—
	2	6.6	3.7	2.6x	—	224	275x	—	110	4.0	—
	3	6.6	3.0	2.8x	—	230	275x	—	111	1.3	—
	4	6.6	3.0	2.9x	—	235	355x	—	114	3.3	—
	5	6.5	2.8	4.1x	—	240	600x	—	110	1.3	—
	6	6.5	3.0	8.6y	—	247	675x	—	122	1.5	—
Radish	1	6.7	3.9	5.2xy	5.6x	297	425x	169x	198	1.5	3.0
	2	6.7	4.0	6.5xy	3.8xy	230	—	226x	190	2.5	2.1
	3	6.7	4.0	3.4x	2.5x	225	350x	125x	200	3.7	2.4
	4	6.7	4.0	3.8x	1.1x	235	370x	100x	106	2.0	1.5
	5	6.6	4.2	3.8x	1.3x	267	442x	124x	127	4.6	2.2
	6	6.6	3.8	8.1y	4.1xy	243	500x	262x	115	2.1	3.5
Tomato	1	6.6	3.6	9.5x	0.3x	290	158x	41	150	2.9	3.8
	2	6.7	3.5	10.7x	0.2x	285	185x	44	150	3.8	2.3
	3	6.6	2.7	9.6x	0.3x	235	185x	96	180	2.2	1.9

*pH of saturated soil paste measured at the end of each cropping.

†Same letter following the plant tissue Cd and Zn content of each plant species indicated the metal content between croppings were not significantly different at 0.05 significant level.

periment, residual Cd and Zn effects were essentially unchanged for the duration. However, it was not known whether residual effects of the sludge-borne metals would be altered with time beyond the duration studied.

Single sludge application

If the soil received a single large dosage of sludge and then was cropped continuously, the plant tissue metal content of each harvested crop would represent the time-dependent plant availability of the soil's heavy metal elements. Under these circumstances, the total metal content of the sludge-affected soil would remain the same throughout the experimental period; however, the metal contents in plant-available forms would change with the chemical condition of the soil after the sludge treatment. Ideally, the amount of sludge added to the soil for this treatment equalled the amount added for the residual treatment. We believed that the increased Cd and Zn content of the composted sludge accounted for most of the higher concentration; however, no attempt was made to compare the outcome of this portion of the experiment with that of the residual experiment.

Statistical analysis of the results indicated that Cd and Zn contents of the harvested

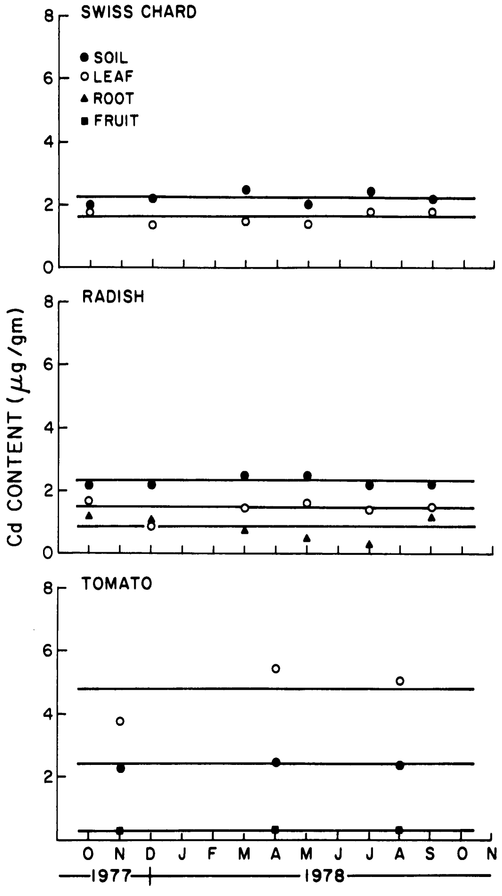

Fig. 3. Plant tissue Cd of vegetable crops grown on the soil (Domino silty clay) where sludge application was terminated.

plant tissue were significantly different between croppings (Table 9 and 10). In many cases, the plant tissue metal contents (Cd and Zn) of the first crop following the sludge application were significantly higher than those in successive croppings. After the first cropping, the Cd and Zn concentrations of harvested plant tissue reduced to a stable level within two to three croppings. There were also several occasions when the Cd and Zn contents of plant tissue of the sixth cropping rose sharply. It was not clear what caused the apparently stabile plant tissue Cd and Zn levels to elevate suddenly. Nevertheless, this fact demonstrates the potential of deposited Cd and Zn to cause metal levels in successive croppings to vary over a wide range. Conversely, the contents of Cd and Zn in harvested plant tissue were sometimes reduced significantly after the first cropping. The resulting metal content of crops always exceeded that of soils not contaminated by sludges.

Table 10. CADMIUM, ZINC, AND LEAD CONTENTS OF VEGETABLES (μ g/gm) HARVESTED FROM A DOMINO LOAM SOIL TREATED WITH A SINGLE SLUDGE INPUT (135 mT/HA) BEFORE CROPPINGS

			Cadmium†			**Zinc†**			**Lead**		
Plant	**Cropping**	**pH***	**Soil**	**Leaf**	**Root/ fruit**	**Soil**	**Leaf**	**Root/ fruit**	**Soil**	**Leaf**	**Root/ fruit**
Swiss chard	1	6.9	4.6	3.2y	—	396	240x	—	150	2.1	—
	2	6.9	4.2	2.6yx	—	330	185x	—	151	5.0	—
	3	6.9	4.5	2.0x	—	350	169x	—	210	1.5	—
	4	7.0	4.0	1.7x	—	340	119x	—	210	3.4	—
	5	6.8	4.6	1.8x	—	321	301x	—	146	2.2	—
	6	6.7	3.9	3.9y	—	327	325x	—	156	1.2	—
Radish	1	6.9	4.2	5.9y	2.9x	346	300x	133x	270	1.9	3.5
	2	7.1	3.8	2.9x	2.3x	325	322x	113x	139	2.5	3.3
	3	7.0	4.0	3.3x	1.9x	330	300x	109x	190	2.8	2.0
	4	7.0	4.0	3.0x	1.0x	390	350x	88x	200	1.5	1.5
	5	7.0	3.8	3.2x	0.8x	335	330x	82x	164	2.7	2.2
	6	6.9	3.6	4.5x	1.8x	332	325x	118x	159	1.7	3.2
Tomato	1	6.9	4.1	8.8x	0.4x	346	120x	36x	148	2.0	3.3
	2	6.9	3.5	7.8x	0.6x	332	125x	41x	170	6.0	1.5
	3	6.9	3.6	5.6x	0.3x	320	100x	32x	160	2.0	1.5

*pH of saturated soil paste measured at the end of each cropping.

†Same letter following the plant tissue Cd and Zn content of each plant species indicated the metal content between croppings were not significantly different at 0.05 significant level.

CONCLUSIONS

This experiment was initiated to examine the pattern of Cd and Zn accumulation in crops grown on sludge-affected soils. Although results are not clear-cut, we demonstrated trends for Cd and Zn accumulation into plant tissue under several different forms of sludge deposition in the soil. Following sludge application, all experimental crops experienced rising Cd and Zn concentrations in the plant tissue. It was theorized that the chemical conditions of the sludge-affected soil would play a key role in the outcome. When soil was treated with sludges, metal content of crops might continue to rise with each application. This would indicate a continuously changing soil chemical condition, yielding more plant-available metals or the metal contents of the crops might remain at equilibrium for several croppings before the repeated sludge applications induced changes in the soil chemical condition. We also observed that the first crop following a sludge application usually would have higher plant-tissue metal contents. If the sludge application was not repeated, metal content of succeeding croppings would decrease to a stabilized level greater than controls. Even for soils on which sludge applications had been terminated for a long time, plant tissue from harvested crops still exhibited elevated levels of metal concentrations in the plant tissue.

It apeared that sludge disposal would have long lasting effects on plant tissue accumulation of heavy metals. Since the diminishing effects of sludge-borne heavy metals in soil were the result of changing soil chemical equilibrium, and since the soil chemical conditions could be changed by repeated sludge applications, it would not be desirable to base the strategy of cropland sludge application entirely on the diminishing heavy-metal effects in the soil. However, it was possible to minimize the hazard associated with the metal accumulation in the soil by proper selection of crops, and by maintaining favorable soil condition.

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