

## Availability of Metals in Irradiated Sewage Sludge

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A variety of vegetables and wheat were grown in pots with a sandy loam soil amended with irradiated, digested sewage sludge or nonirradiated, digested sewage sludge to determine difference in availability of Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn, K, Ca, and Mg. Irradiation of the sludge with high-energy electrons for disinfection did not change the availability of the metals. Concentration of metals in plants treated with irradiated, digested sludge was similar to that of metals in plants treated with nonirradiated, digested sludge. Plants grown with both irradiated and nonirradiated sludge should be monitored to ensure that high levels of metals do not accumulate in the plants.

Irradiation of municipal sludge for disinfection may prove to be the most important new treatment method that has emerged in the last half century (National Science Foundation, 1976). Work has shown that sludges irradiated with both radioactive sources (Suess et al., 1975; McCaslin and Titman, 1977) and high-energy electrons (Kirkham and Manning, 1979) can be used agriculturally. Even though much is known about the availability of toxic heavy metals in nonirradiated sludges (Page, 1974; Council for Agricultural Science and Technology, 1976; Kirkham, 1977), no study has been done which compares the availability of heavy metals of concern in irradiated sludge to that of metals in nonirradiated sludge. Theoretical considerations do not favor the possibility that the chemical behavior of the inorganic heavy metals would be changed by the irradiation treatment, which occurs at the treatment plant and before the sludge is applied to soil. However, the problem is of sufficient importance to demand specific validation because municipalities are installing irradiation facilities. The Deer Island Sewage Treatment Plant of Boston, MA, has a pilot plant that is irradiating sludge (Trump et al., 1977). Miami-Dade Water and Sewer Authority in Florida is planning to irradiate sludge and spread it on land (Williams, 1979). Sludge from Los Angeles, CA, Albuquerque, NM, and Beltsville, MD, is being irradiated for evaluation purposes (Clark, 1979). To safeguard food supplies grown with irradiated sludge, it is important to know if the irradiation treatment changes metal availability. Therefore, the objective of this research was to determine if concentration of metals in plants grown with irradiated sludge was different from that in plants grown with the same type of sludge that had not been irradiated.

### EXPERIMENTAL SECTION

Seeds of 10 plant species were planted in 10-cm diameter plastic pots on 22 January 1978 (day 1). The 10 species and number of seeds planted per pot were as follows (common name, scientific name and variety, seeds per pot): bush bean, *Phaseolus vulgaris* L. "Tendergreen Bush", 3; carrot, *Daucus carota* L. "Danvers Half Long", 40; sweet corn, *Zea mays* L. "Tendermost Hybrid", 6; dill, *Anethum graveolens* L. "Mammoth", 30; mustard, *Brassica juncea* Coss. "Florida Broad Leaved", 80; onion, *Allium cepa* L. "Green Bunching", 40; pea, *Pisum sativum* L. "Little Marvel", 5; radish, *Raphanus sativum* L. "Hailstone", 25; tomato, *Lycopersicon esculentum* Mill. "Yellow Jubilee", 14; wheat, *Triticum aestivum* L. em. Thell. "Ponca", 15. These plants were chosen because they are common vegetable and field crops. The soil was a fine sandy loam

Table I

treatment	application rate
no sludge or inorganic fertilizer	50 mL of tap water added per pot on days 22, 25, 27
a 3740-ppm solution of an inorganic fertilizer (Miracle-Gro, Stern's Nurseries, Inc., Geneva, NY; 18% N, 18% P, 21% K, 0.50% Mg, 0.05% Cu, 0.10% Fe, 0.05% Mn, 0.05% Zn)	50 mL added per pot on days 22, 25, 27
digested, nonirradiated sludge	50 mL added per pot on days 22, 25, 27
digested, irradiated sludge	50 mL added per pot on days 22, 25, 27

(Ultic Haplustalf) with 21% silt, 73% sand, 6% clay, a pH of 6.3, and 5  $\mu\text{g/g}$  of  $\text{NO}_3\text{N}$ . It contained, in micrograms/gram, the following amounts of dilute-acid (0.05 N HCl and 0.025 N  $\text{H}_2\text{SO}_4$ ) extractable elements (Isaac and Kerber, 1971): K, 56; Ca, 1018; Mg, 304; Fe, 21.2; Zn, 4.2; Mn, 19.2. The pots were filled to within 2.5 cm of the top with soil.

Two types of sludge were sent in a frozen state to Oklahoma State University from the Deer Island Sewage Treatment Plant of Boston, MA. One sludge was a digested sludge. The other sludge was a digested sludge which had been irradiated with an electron dose of 400 krad. The irradiation process is described by Trump et al. (1977). Both sludges had about 5% solids on a dry weight basis. The digested, nonirradiated sludge and the digested, irradiated sludge contained, in micrograms/gram of dry matter, the following amounts of dilute-acid extractable elements, respectively (Isaac and Kerber, 1971): K, 1124, 3691; Ca, 806, 1030; Mg, 570, 1010; Fe, 140, 145; Mn, 6.9, 4.0; Zn, 44, 56; Cu, 34, 27; Ni, 33, 90; Pb, 45, 66; Cd, 6, 10; Cr, 12, 33. Coefficient of variation for the analysis of the elements in the sludge was 32%. Extractable amounts in the irradiated sludge were similar to those in the nonirradiated sludge.

There were four treatments in which sludge, inorganic fertilizer, or only water were applied (Table I). The tap water contained in micrograms/milliliter the following: Zn, 0.01; Cu, 0.04; Cd, <0.001; Cr, <0.001; Ni, <0.001; Pb, <0.001; Mn, <0.001. There were three pots per treatment for a total of 144 pots (12 plant species  $\times$  4 treatments  $\times$  3 replications). Between days 29 and 56 (harvest), 50 mL of tap water was added each day to the pots. The pots had drainage holes, but water did not usually drain out of the bottom after irrigations. The 150 mL total sludge added to pots that received sludge corresponded to about 9.3 mt/ha (4.1 T/A) sludge on a dry-weight basis. This amount was added because it is a moderate rate of ap-

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Table II. Dry Weight of Shoots and Concentration of Elements in Leaves of Ten Plant Species Grown for 56 Days in 10-cm Diameter Pots with a Fine Sandy Loam Soil Treated with Nonirradiated Digested Sludge, Irradiated Digested Sludge, Inorganic Fertilizer, or Only Water

plant	nonirrad	irrad	inorg fert	water	nonirrad	irrad	inorg fert	water	
		Dry Weight, g/pot					Cadmium (Normal Range: 0.1-5 µg/g), µg/g		
bean	1.38a <sup>b</sup>	1.13a	1.68a	1.71a	0.23b	<0.1a	<0.1a	<0.1a	
carrot	0.45a	0.55a	1.52a	0.46a	2.7a	2.8a	1.8a	2.3a	
corn	1.39a	1.66a	0.84a	1.45a	1.2a	2.3b	1.1a	1.2a	
dill	0.37a	0.28a	1.07a	0.48a	5.4a	c	2.8a	2.9a	
mustard	1.76a	0.68a	4.07b	1.00a	3.3b	2.1b	1.2a	1.2a	
onion	0.17a	0.05a	0.04a	0.05a	0.7a	2.5a	5.0a	4.5a	
pea	c	0.92a	1.48b	0.84a	c	3.3a	1.6a	1.2a	
radish	0.79a	0.86a	1.58a	0.91a	5.6c	4.0c	0.7a	1.2b	
tomato	1.64b	1.89b	1.92b	0.77a	7.2c	7.7c	0.9b	0.7a	
wheat	c	1.06b	2.02c	0.64a	c	2.1a	1.2a	1.4a	
		Calcium (Normal Range: 0.5-5%), %					Chromium (Normal Range: 0.1-15 µg/g), µg/g		
bean	0.94a	0.95a	1.12a	1.45a	15.2ab	4.7b	1.2b	0.2a	
carrot	1.29a	1.21a	1.08a	1.27a	44.4b	86.1b	4.2a	9.3a	
corn	0.48a	0.42a	0.23a	0.38a	6.1a	5.1a	1.4a	3.0a	
dill	c	c	0.86a	1.65b	83.5c	c	4.1b	<0.1a	
mustard	1.46a	1.53a	1.44a	1.64a	67.7b	37.8b	2.1a	1.6a	
onion	0.90a	0.96a	0.78a	0.78a	25.2a	17.5a	4.1a	3.3a	
pea	c	1.21a	1.38a	1.74a	c	100.86	5.4a	1.4a	
radish	1.61a	1.64a	1.44a	1.70a	161.0b	98.2b	4.0a	3.5a	
tomato	1.63a	1.51a	1.44a	2.45b	157.3b	144.9b	5.8a	4.8a	
wheat	c	0.50a	0.55a	0.66a	c	11.4b	2.1b	0.2a	
		Copper (Normal Range: 4-40 µg/g), µg/g					Iron (Normal Range: 25-300 µg/g), µg/g		
bean	21.0b	16.6b	15.2b	7.9a	169a	148a	222a	201a	
carrot	43.1c	72.1c	16.8b	4.2a	264a	361a	291a	310a	
corn	12.4b	13.8b	19.3b	7.7a	240a	117a	129a	154a	
dill	56.5c	c	5.9b	4.0a	525a	c	167a	305a	
mustard	59.3b	44.8b	9.8a	16.3a	449a	433a	366a	411a	
onion	14.0a	12.5a	16.9a	9.0a	99a	315a	307a	350a	
pea	c	79.1c	13.5b	7.5a	c	210b	234b	166a	
radish	162.8c	111.5bc	40.8b	8.9a	844b	573b	197a	188a	
tomato	97.8b	102.0b	16.1a	11.4a	420b	316a	317a	299a	
wheat	c	20.1c	9.1b	4.9a	c	199a	190a	261a	
		Lead (Normal Range: 0.1-45 µg/g), µg/g					Magnesium (Normal Range: 0.1-1%), %		
bean	9.3a	14.7a	14.9a	16.8a	0.15a	0.15a	0.15a	0.16a	
carrot	31.0a	23.1a	32.9a	30.3a	0.25a	0.15a	0.15a	0.20a	
corn	13.8a	10.7a	10.5a	11.9a	0.16a	0.16a	0.14a	0.16a	
dill	0.1a	c	20.6a	43.8a	0.27a	c	0.20a	0.28a	
mustard	23.1a	22.4a	18.7a	23.3a	0.16a	0.16a	0.16a	0.16a	
onion	4.9a	37.5a	11.3a	13.0a	0.12a	0.12a	0.17a	0.17a	
pea	c	24.5a	24.7a	35.2a	c	0.16a	0.16a	0.16a	
radish	29.9a	29.2a	15.4a	14.2a	0.20a	0.16a	0.16a	0.16a	
tomato	26.4b	20.1ab	11.9a	12.4ab	0.17a	0.17a	0.16a	0.18a	
wheat	c	10.3a	19.1a	16.3a	c	0.15a	0.15a	0.16a	
		Manganese (Normal Range: 25-300 µg/g), µg/g					Nickel (Normal Range: 0.1-50 µg/g), µg/g		
bean	117a	84a	83a	104a	4.0a	4.9a	4.7a	3.3a	
carrot	79a	222a	64a	68a	6.7a	11.9a	4.2a	7.0a	
corn	67c	55bc	45ab	38a	2.3a	1.9a	1.8a	2.3a	
dill	81a	c	43a	120a	5.4a	c	1.1a	5.8a	
mustard	95b	99b	53a	74b	8.9b	6.3b	4.2a	4.7a	
onion	15a	115a	96a	63a	3.5a	15.0a	8.5a	14.0a	
pea	c	173a	115a	138a	c	5.1a	4.0a	1.6a	
radish	97c	80bc	48ab	40a	18.2c	12.4b	4.4a	4.7a	
tomato	189b	186b	66a	84a	13.5b	17.0ab	3.7a	4.1a	
wheat	c	119b	78a	141b	c	1.6a	0.9a	3.7a	
		Potassium (Normal Range: 0.1-5%), %					Zinc (Normal Range: 10-400 µg/g), µg/g		
bean	0.18b	0.18b	0.18b	0.17a	54.6b	39.7ab	35.7a	22.2a	
carrot	0.30a	0.17a	0.18a	0.28a	214.0c	149.8c	55.3b	30.3a	
corn	0.18a	0.18a	0.18a	0.13a	110.4b	140.9b	93.1b	30.1a	
dill	0.65b	c	0.37a	0.38a	234.2b	c	40.2a	67.1a	
mustard	0.18a	0.18a	0.18a	0.19a	121.1b	108.5b	41.3a	50.9a	
onion	0.15a	0.62a	0.74a	0.85a	31.5a	62.5a	50.8a	56.3a	
pea	c	0.17b	0.18b	0.17a	c	171.7c	42.0b	19.8a	
radish	0.25b	0.18b	0.18ab	0.18a	297.1c	197.2c	63.2b	40.1a	
tomato	0.17a	0.18b	0.18b	0.25b	211.9c	221.2c	26.1b	13.7a	
wheat	c	0.15a	0.17a	0.16a	c	43.9b	32.7b	17.0a	

<sup>a</sup> Normal range in plants determined using values published by Chapman (1973), Kirkham (1975, 1977), and Zimdahl and Koeppel (1977). <sup>b</sup> Means in each row followed by the same letter are not significantly different at the 0.05 level according to Duncan's new multiple range test. <sup>c</sup> Insufficient sample for analysis.

plication. Sludge was applied after planting, as is done under field conditions when liquid sludge is spread on established crops.

The experiment was carried out in a growth room. The day and night temperatures varied from 21 to 26 °C and 15 to 20 °C, respectively. The relative humidity varied from 40 to 54%. The average pan evaporation rate during the 44-day experiment was 0.30 cm day<sup>-1</sup>. The flux density of incident light, provided by cool white fluorescent lamps, was 300 μE m<sup>-2</sup> s<sup>-1</sup> from 0600 to 1800 h.

Plant height was measured on days 20, 27, 34, 41, 49, and 56. On day 56, the number of plants in each pot was counted, plants were cut above the soil or sludge surface, and plant tops were dried at 70 °C. After drying, dry weight was determined. Plants were ground with a porcelain mortar and pestle and analyzed, using a nitric-perchloric acid digest and an atomic absorption spectrophotometer, for K, Mg, Ca, Fe, Mn, Zn, Cu, Ni, Cd, Cr, and Pb (Isaac and Kerber, 1971).

Results presented are the averages of three values (three replications). Data were analyzed statistically using Duncan's new multiple-range test (Steel and Torrie, 1960).

#### RESULTS AND DISCUSSION

Irradiation had little effect on availability of heavy metals to plants (Table II). Concentrations of heavy metals in crops grown with irradiated, digested sludge were similar to those in crops grown with nonirradiated, digested sludge, except for four instances: Cd in bean and corn, Fe in tomato, and Ni in radish. There was no consistent difference for these four exceptions. Cadmium in bean grown with nonirradiated sludge was more than Cd in bean grown with irradiated sludge. But Cd in corn grown with irradiated sludge was more than Cd in corn grown with nonirradiated sludge.

High metal concentrations were observed in some of the sludge-treated plants. Except for corn and wheat, Cr was high in all plants grown with sludge. Cadmium was high in dill, radish, and tomato and Cu was elevated in carrot, dill, mustard, radish, and tomato grown with sludge. High levels of metals in sludge-treated plants, particularly the leafy parts of vegetables, have been noted in many other experiments (Page, 1974; Council for Agricultural Science and Technology, 1976; Kirkham, 1977).

Table II also shows the dry-weight results. Height results are not shown but paralleled dry-weight results. Height results are available (Kirkham, 1977-1978). At harvest, there was no difference in growth among the treatments except for the following: mustard, pea, tomato, wheat. Mustard, pea, and wheat grown with inorganic fertilizer produced more dry weight than did the same plants grown with sewage sludge or with no fertilizer. Tomatoes grown with both sewage sludge and inorganic fertilizer grew better than tomatoes which were not fertilized. Concentration of K, Ca, and Mg in plants grown with irradiated, digested sludge was similar to that in plants grown with digested, nonirradiated sludge (Table II).

In another study (Kirkham, 1977-1978), plants were grown with irradiated, raw sludge and nonirradiated, raw sludge instead of irradiated, digested sludge and nonirradiated, digested sludge. In both studies, the same plant varieties were grown and plants were analyzed for K, Ca, Mg, Fe, Zn, Cu, Ni, Cr, and Cd. Plants grown with raw sludge (irradiated or nonirradiated) had concentrations of K, Ca, and Mg higher than plants grown with digested sludge (irradiated or nonirradiated). Also, plants grown

with digested sludge (irradiated or nonirradiated) usually had higher concentrations of Fe, Zn, Cu, Ni, Cr, and Cd than did plants grown with raw sludge (irradiated or nonirradiated). It has been suggested that raw sludge could be irradiated for disinfection and injected into the soil (Smith et al., 1977; Trump et al., 1977). The lengthy digestion process would not be necessary for disinfection. If raw sludge were irradiated and used on agricultural land, concentrations of metals in plants might be lower than if digested, nonirradiated sludge were used.

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