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TRANSFER OF COBALT-60 TO PLANTS FROM SOILS TREATED WITH SEWAGE SLUDGE

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Abstract. The uptake of ^{60}Co from soils fertilized with contaminated sewage sludge has been investigated under a variety of experimental conditions. A number of garden plots were prepared by thoroughly mixing sludge containing ^{60}Co with farm soils from the Ottawa Valley. Edible plants were grown in the open on these plots under conditions approximating those in market gardens. The crops were harvested at maturity and were prepared for measurement of ^{60}Co by drying portions of the roots, leaves, stems and fruit.

The samples were counted on a large germanium detector which was capable of resolving ^{60}Co from other gamma-emitting nuclides. Cobalt was readily taken up from contaminated sludge but was nonuniformly distributed in various parts of the plant. In general, the roots showed the highest levels while edible portions such as seeds and tubers had much lower concentrations. The uptake ratio, expressed as radioactivity in the sample to radioactivity in the soil, varied from 0.003 to 8 on a dry-weight basis.

Key words: Cobalt uptake; sludge; plant uptake; cobalt-60; uptake ratio; radioactivity in plants; soil transfer.

INTRODUCTION

Numerous investigators have demonstrated the beneficial effects of sludge when applied regularly to the soil (Anderson 1955, Kelling et al. 1974); some of these authors also have considered deleterious effects such as the uptake of heavy metals from certain types of waste sludge (Black and Kronis 1974). These effects are particularly important in Ontario where many towns dump the solid wastes on sanitary landfills or spread them on nearby farmland. The removal of phosphate from waste streams with lime or other chemical precipitants will, in fact, cause sludge to be more valuable as fertilizer. Although such methods will coprecipitate trace elements such as cobalt, the higher pH of the resulting lime-sludges should reduce the availability of heavy metals through the formation of insoluble bases in the soil.

Radioactive isotopes such as ^{60}Co are being used in industry and medicine in increasing amounts. This increases the chance that the nuclide will appear in the sludge produced by city waste-treatment plants.

Sludge

Analyses of sludge from one of the Chalk River Nuclear Laboratories (CRNL) waste-treatment systems showed that ^{60}Co was distributed fairly uniformly throughout the solids at a concentration of 50 pCi/g dry-weight. Preliminary measurements on tomato plants which germinated spontaneously at the drainage sump showed measurable transfer to edible parts of the plant, so it was decided to make a more detailed study of the soil-plant transfer of the radionuclide.

At CRNL the waste streams are carefully divided. Reactor cooling water, active drains, storm drains, storm sewers and sanitary sewers are discharged separately, but small amounts of ^{60}Co from laboratory areas are always present in the sanitary sewage. Recovery of the solids by primary settling (Imhoff tank) of the sanitary sewage is followed by sand filtration (lagooning) to reduce the water content and increase solids to 16% on a dry-weight basis. About half (30-70%) of the cobalt is normally found associated with the solids following settling, the remainder being discharged with the liquid wastes.

In the following experiments contaminated sludge from CRNL was used in combination with three different types of soils to estimate the plant uptake of ^{60}Co under various growing conditions.

METHODS

Crops were grown under field conditions in rectangular plots. The plots were enclosed by wooden frames 155 x 205 x 24 cm, which were open at the bottom and sunk to a depth of 20 cm in a fenced area. Three parts soil to one part sludge, by volume, were mixed separately; tracer was added if required, by treating the soil mixture with a dilute aqueous solution containing ^{60}Co and the other isotopes which were being studied.

The treated soil was then thoroughly mixed in a plastic bag and placed loosely in the frames to a depth of 20 cm.

Table 1 lists the chemical properties of various types of soil used in the experiments. Grenville loam is a well-drained Brown Forest soil from the Ottawa Experimental Farm (Agriculture Canada); CRNL loam is an unclassified, sandy, well-drained forest soil from the Chalk River area; Acid Peat, formed from decayed sphagnum moss, is from the same area.

Two different forms of radioactive cobalt were used. The sludge itself contained sufficient ^{60}Co for uptake experiments provided that relatively high application rates were used. This cobalt is presumably bound as a complex with the organic sludge and should be less available for uptake by plants than ^{60}Co in solution. For comparison, a solution (0.01 mℓ) of high specific activity $^{60}Co^{++}$ was diluted with 50 ℓ of Ottawa River water and used as a tracer on half the plots. The amount of ^{60}Co added (200-500 pCi/g soil) was sufficient to override the ^{60}Co already present in the sludge (6-50 pCi/g). Identical plots were prepared in triplicate and were seeded in late spring with the species under study. In the case of tomatoes, small seedlings were used because of the relatively short growing season at Chalk River (90-100 days).

Plants were harvested at maturity, then weighed, dried at 105°C, reweighed and counted. Roots and tubers were carefully washed and scrubbed with a brush before drying. In most instances the skin was peeled from below ground parts and treated separately. Cobalt-60 and other nuclides including ^{137}Cs, ^{85}Sr, ^{40}K, ^{65}Zn and ^{54}Mn were measured simultaneously on a low background Ge(Li) gamma spectrometer. This system has previously been described in detail elsewhere (Grummitt 1971). Results for uptake of elements other than cobalt will not be discussed here.

RESULTS AND DISCUSSION

Every effort was made to grow the same species of plant on each type of soil, but poor germination and damage by insects and rodents sometimes resulted in insufficient material being available for replicate analysis. Nevertheless, as over 300 samples were analyzed, the individual results are too numerous to discuss in detail and will be made available in a separate report.

The yield of oats grown on Acid Peat was very low compared with mineral soils (Table 2). Evans and Dekker (1965) reported a similar effect for 100% peat. The addition of sludge to CRNL loam increased the yield of oats appreciably but did not alter it for potatoes and tomatoes. Tomatoes, however, showed a similar yield increase on Acid Peat with sludge. In general, the addition of sludge had minimal effect on crop yields and showed possible toxic effects only at the 100% concentration.

One of the most interesting features of the results was the great variability in uptake ratios (^{60}Co/g of dry tissue: ^{60}Co/g of dry soil) when different species were grown under identical conditions (Table 3). Crossley (1969) found in a field study at Oak Ridge, Tennessee, that ^{60}Co concentrations in vegetation showed no regular patterns and were unrelated to measured soil concentration. He attributed this to inadequate soil sampling. Hill et al. (1953) measured the stable cobalt in crops and weeds grown under uniform soil conditions. Five cereals varied from 0.01 to 0.7 μg/g. The authors concluded that the nature of the plant was the most important factor in determining cobalt content.

In the present case similar species differences were observed. Cobalt-60 concentrations were an order-of-magnitude lower in corn stover than in potato stems and leaves from the same plot; uptake by beans was greater than corn by a factor of 80 ± 6.6. These differences were significant at the

TABLE 1. Chemical properties of sludge and soils. The weight of replaceable ion is given per gram of air-dried soil. Values in μg/g x 2 give lb/acre.

	pH	Phosphorus P_2O_5	Potassium K_2O	Calcium CaO	Magnesium MgO	Cobalt Co
CRNL (unclassified)	5.8	130 μg/g	46 μg/g	230 μg/g	23 μg/g	1.1 μg/g
Grenville loam	7.4	-	240	3800	350	1.4
Acid Peat	5.1	-	70	6600	710	0.61
Sewage Sludge	4.6	-	170	380	140	3.7

TABLE 2. Yields of crops grown on various soils.

	Oats	Potatoes	Tomatoes
CRNL loam	160 kg/ha	2300 kg/ha	2700 kg/ha
CRNL + sludge	310	2500	2500
Grenville loam	210	780	2600
Grenville + sludge	200	560	2700
Acid Peat	60	1200	200
Peat + sludge	60	1100	500
100% sludge	20	900	1100

0.1% level. Significant differences were evident for most of the species in this study suggesting that, as in the case of stable cobalt, the variations were probably genetically controlled.

Various parts of the plant showed concentrations differing by an equally large factor (Table 3). The uptake by corn leaves was 20.0 ± 1.0 times higher than that by the grain. For oats the differences were smaller, the average stover/grain ratio for all soils being 2.4 ± 0.5 (Table 5). In both cases the differences were significant at the 1% level.

Highest concentrations generally were observed in the rooting parts of the plant. In at least half the cases the uptake ratio lay in the range 1.5 to 6.6. Values greater than unity cannot be caused by adhesion of soil particles and therefore must be due to some effect such as adsorption on the large surfaces of the rootlets.

Factors relating to the growing conditions of a particular crop do not have as large an effect on the uptake ratio as does the species of plant. In Table 4 the results of liming two acid soils is shown for a variety of crops. Liming decreases the mean cobalt uptake by a factor of 2.7 ± 0.4, the effect being in the expected direction. Analysis of variance of the data shows the difference to be significant at the 0.1% level. The smaller uptake from Grenville versus CRNL loam (Tables 3 and 5) is partially due to the much higher pH of the Grenville. Unfortunately no data are available comparing uptake from these two loams at the same

TABLE 3. Plant uptake of ^{60}Co. Effect of the addition of 300 t/ha sludge to three types of soil (with S. E.).

Soil type	Plant species	Initial pH	Uptake ratios: No sludge	Uptake ratios: 300 t/ha sludge
CRNL loam	Radish, edible portion	5.0	1.3 ± 0.3	0.25 ± 0.03
	Radish, leaves	5.0	0.84 ± 0.15	0.47 ± 0.03
CRNL loam	Oats, grain	5.0	0.06 ± 0.01	0.02 ± 0.004
	Oats, stover	5.0	0.25 ± 0.03	0.04 ± 0.006
CRNL loam	Corn, grain	5.0	0.008 ± 0.001	0.005 ± 0.001
	Corn, stover	5.0	0.030 ± 0.002	0.021 ± 0.004
	Corn, leaves	5.0	0.16 ± 0.020	0.050 ± 0.001
CRNL loam	Snap Beans	5.0	0.62 ± 0.02	0.30 ± 0.02
	Snap Beans, stem & leaves	5.0	0.23 ± 0.02	0.10 ± 0.01
CRNL loam	Potatoes, tubers	5.0	0.12 ± 0.01	0.10 ± 0.02
	Potatoes, stem & leaves	5.0	1.2 ± 0.08	2.1 ± 0.08
CRNL loam	Tomatoes, fruit	5.0	0.07 ± 0.02	0.09 ± 0.03
	Tomatoes, stem & leaves	5.0	0.67 ± 0.10	0.29 ± 0.09
CRNL loam	Carrots, edible portion	5.0	0.18 ± 0.02	0.14 ± 0.02
	Carrots, tops	5.0	0.32 ± 0.09	0.14 ± 0.003
Acid Peat	Radish, edible portion	6.6	0.43 ± 0.08	0.07 ± 0.02
	Radish, leaves	6.6	0.55 ± 0.18	0.23 ± 0.06
Grenville loam	Radish, edible portion	7.4	0.23 ± 0.04	1.0 ± 0.18
	Radish, leaves	7.4	0.71 ± 0.20	1.2 ± 0.30
Grenville Loam	Potatoes, tubers	7.4	0.03 ± 0.003	0.27 ± 0.008
	Potatoes, stem & leaves	7.4	1.2 ± 0.36	1.8 ± 0.07

TABLE 4. Effect of pH on ^{60}Co uptake ratios (with S.E.). The mean uptake ratios excluding 100% sludge, acid/limed = 2.7 ± 0.4.

Soil type	Sludge	Plant species	pH Initial	pH Limed	Uptake ratios Acid	Uptake ratios Limed	Acid/Limed
CRNL loam	300 t/ha	Radish, edible portion	5.0	6.5	0.25 ± 0.03	0.050 ± 0.01	5.0
CRNL loam	300 t/ha	Radish, leaves	5.0	6.5	0.47 ± 0.03	0.32 ± 0.05	1.5
CRNL loam	300 t/ha	Potatoes, tubers	5.0	6.5	0.10 ± 0.02	0.054 ± 0.008	1.9
CRNL loam	300 t/ha	Potatoes, stem & leaves	5.0	6.5	2.1 ± 0.06	0.70 ± 0.15	3.0
Acid Peat	0	Potatoes, tubers	5.1	6.6	0.08 ± 0.01	0.040 ± 0.005	2.0
Acid Peat	0	Potatoes, stem & leaves	5.1	6.6	0.81 ± 0.07	0.36 ± 0.03	1.4
Acid Peat	0	Parsnips, edible portion	5.1	6.6	0.13 ± 0.01	0.043 ± 0.003	3.0
Acid Peat	0	Parsnips, leaves	5.1	6.6	0.19 ± 0.02	0.054 ± 0.004	3.5
100% sludge	1200 t/ha	Potatoes, tubers	4.6	6.4	0.05 ± 0.01	0.05 ± 0.01	1.0
100% sludge	1200 t/ha	Potatoes, stem & leaves	4.6	6.4	0.57 ± 0.18	1.4 ± 0.3	0.4

TABLE 5. Uptake of ^{60}Co by oats grown in various soils (with S.E.).

Soil type	Sludge	Plant species	Uptake ratio
CRNL loam	nil	Oats, grain	0.06 ± 0.01
CRNL loam	300 t/ha	Oats, grain	0.02 ± 0.004
Grenville	nil	Oats, grain	0.04 ± 0.004
Acid Peat	nil	Oats, grain	0.04 ± 0.003
No mineral soil added (100% sludge)	1200 t/ha	Oats, grain	0.38 ± 0.02
CRNL loam	nil	Oats, stover	0.25 ± 0.03
CRNL loam	300 t/ha	Oats, stover	0.04 ± 0.006
Grenville	nil	Oats, stover	0.04 ± 0.003
Acid Peat	nil	Oats, stover	0.09 ± 0.007
No mineral soil added (100% sludge)	1200 t/ha	Oats, stover	1.01 ± 0.08

pH. One of the more surprising results is that liming the 100% sludge plot had little effect on cobalt uptake, e.g. the ratio for potato tubers, limed/unlimed, is 1.0 ± 0.14. No explanation for this behavior can be offered.

Cobalt can form stable compounds with organic matter and so become relatively unavailable for uptake by plants. Tables 3 and 5 show the effect of addition of sludge to mineral and organic type soils. The addition of sludge to CRNL loam and to peat reduces availability by 0.54 ± 0.08, the effect being significant at the 0.1% level. In the case of Grenville loam the results appear to be reversed, the increase being by a factor of 4.1 ± 1.8 (significant at the 5% level).

For CRNL loam the uptake of ^{60}Co added in sludge was 2.0 ± 0.4 times higher than when tracer $^{60}Co^{++}$ was added. In contrast, with 100% sludge, tracer increased the uptake ratios by a mean factor of 5.0 ± 1.3 (Table 6). These values are significant at the 0.1% and 1% level, respectively. The results suggest that $^{60}Co^{++}$ added to mineral soil is fixed so that it is less readily available to plants.

In Table 7, mean uptake ratios are given for the edible portion of various field crops which form a part of man's diet in North America. Values are also expressed on a wet-weight basis to facilitate calculating the transfer of ^{60}Co from raw sewage to diet, assuming the contaminated sludge is used as a fertilizer on farmland. Except for corn grain, the uptake of ^{60}Co is fairly similar in all of the plants studied (range 0.01 to 0.04 expressed on a wet-weight basis).

CONCLUSIONS

The uptake of ^{60}Co by plants from soils fertilized with sewage sludge has been studied in detail. Factors such as pH, type of soil, addition of sludge and availability of ^{60}Co have a relatively small effect on the uptake ratios. On the other hand, the ^{60}Co content varies greatly with plant species and with various parts of the plant. Mean uptake ratios for a number of edible plants are in the range 0.01 to 0.04 on a wet-weight basis. These data should assist in calculating the transfer of ^{60}Co from sewage sludge to man.

TABLE 6. Availability of various forms of cobalt (with S.E.).

Soil type	Plant species	pH	Uptake ratios	
			^{60}Co from sludge	^{60}Co tracer added
CRNL loam + 300 t/ha sludge	Snap Beans	5.0	0.30 ± 0.02	0.29 ± 0.02
	Snap Beans, stem & leaves	5.0	0.15 ± 0.01	0.10 ± 0.01
	Tomatoes, fruit	5.0	0.25 ± 0.03	0.09 ± 0.01
	Tomatoes, stem & leaves	5.0	0.89 ± 0.05	0.20 ± 0.03
	Potatoes, tubers	6.5	0.06 ± 0.01	0.05 ± 0.008
	Potatoes, stem & leaves	6.5	0.94 ± 0.06	0.45 ± 0.02
100% sludge	Potatoes, tubers	6.4	0.05 ± 0.01	0.18 ± 0.004
	Potatoes, stem & leaves	6.4	1.4 ± 0.3	4.2 ± 0.9
	Tomatoes, fruit	4.6	0.18 ± 0.01	0.86 ± 0.05
	Tomatoes, stem & leaves	4.6	0.6 ± 0.06	5.2 ± 0.3

TABLE 7. Mean uptake ratios for edible food.

	Uptake ratio[a]	Wet-weight uptake ratio[b]
Cereals		
Oats	0.10	0.015
Corn	0.008	0.003
Root Crops		
Radish	0.55	0.032
Carrots	0.12	0.011
Parsnips	0.12	0.016
Turnips	0.25	0.043
Legumes		
Snap Beans	0.41	0.010
Tubers		
Potatoes	0.06	0.020
Fruit		
Tomatoes	0.25	0.011

[a]pCi/g dry plant tissue/pCi/g dry soil.
[b]pCi/g plant wet-weight/pCi/g dry soil.

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