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Sewage Sludge as a Source of Cadmium in Soil–Plant–Animal Systems

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The objective of this presentation is to relate the abundance and mobility of Cd in components of terrestrial ecosystems with implications for land utilization of sewage sludge. The uptake of Cd by crop plants is a function of the quantity of the element in the soil plus other soil factors affecting the Cd ion activity or electrochemical potential at the plant root surface. The natural abundance of Cd in soils has been reported as 0.5 μg/g which is higher than the background level of 0.2 μg/g found in soils studied in Pennsylvania. Experimental results indicate that the plant availability of Cd increases with each soil addition. While the plant availability of Cd is decreased by liming to increase soil pH, it has not been possible to add Cd salts or sewage sludge Cd without significantly increasing plant uptake.

Field studies have shown that land application of sewage sludge can be expected to increase the Cd concentration of corn leaves from a range of 0.05–0.1 μg/g to 1–3 μg/g. Two years after the last application of sludge which added up to 10 ppm Cd to the surface soil, corn grain, sorghum grain, wheat grain, and potatoes showed a 10- to 15-fold increase in Cd over background levels. Studies were conducted with chicks, laying hens, and meadow voles (*Microtus Pennsylvanias*) to assess the impact of this increase in plant Cd upon the food chain.

Corn and sorghum plants were grown on soils with either inorganic or sludge fertilizer for the purpose of producing herbage for use in feeding trials with meadow voles. Eight diets and a synthetic control diet were formulated to study the effect of source (plant vs. inorganic) of Cd on tissue accumulation. Significant accumulation of Cd occurred in kidney and liver, but not muscle, of voles fed diets containing sludge fertilized corn (1.09 μg/g) or sludge fertilized sorghum (2.76 μg/g). The source of Cd had little influence on tissue accumulation.

In studies with broiler chicks and laying hens, natural diets containing 0.2 ppm Cd were supplemented with 3 ppm of this element. As with the meadow voles, Cd readily accumulated in liver and kidney. Although the results were not statistically significant, 3 ppm dietary Cd doubled muscle Cd content. There was no transfer of Cd to egg in a long term (12 month) experiment with laying hens.

Soil management programs have been developed to maintain animal dietary levels of Cd at less than 1.0 μg/g from the use of sewage sludge on land in Pennsylvania. However, it is concluded that this level over time may cause a significant accumulation of Cd in animal tissues. Interpretation of these results in relation to those for human intake of Cd and the long range health effects of Cd is required for the proper monitoring of sewage sludge applications on land used for production of crops which enter the food chain.

Introduction

The accumulation of Cd and other ions by plants from soil is affected by inherent differences among species and varieties within species, soil abundance of the element, ionic interactions in soils, and soil-plant interactions. Interpretations of experimental results relating soil testing, soil chemical equilibria, plant analysis, and plant physiological processes have been subjects of several reviews (*1–9*). The review on trace element cycling by Allaway in 1968 (*10*) provides an excellent discussion of principles. Bingham et al. (*11*) reported that additions of 4 to 640 ppm Cd to soil resulted in a 25% decrement in plant yield, diagnostic leaf concentrations of 3 to 160 ppm Cd, and concentrations within the edible portions of plants ranging from 2.0 to 80 ppm Cd, depending upon plant species. The relatively high concentrations of Cd found toxic to plants when compared to concentrations of Cd considered harmful to animals and man make results of plant toxicity studies seem irrelevant to the problem of Cd in components of terrestrial ecosystems. The objective of this presentation is to review experimental results from several investigations conducted at Penn State University that have led to a

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chemical monitoring program designed to prevent excessive food chain levels of Cd from the utilization of sewage sludge as fertilizer on land used to produce food chain crops. The suitability of sludge for use on cropland is determined by the Cd content and the rate of sludge applied to supply the required N and P.

Soils and Soil–Plant Relationship

From results summarized by Baker and Chesnin (*2*), the abundance of Cd increases from an average of 0.13 ppm in igneous rocks to 0.18 ppm for the average in the lithosphere to 0.5 ppm as an average for soils. The range of Cd in virgin soils has been reported as 0.01 to 0.7 ppm. The field soil used for the Penn State studies was on a land boundary between Hagerstown silt loam and Murrill silt loam. The total Cd in the soil, sampled from different areas of the field, ranged from 0.2 to 0.3 ppm. Thus, the plants were established on plots with a relatively low background level of Cd. The details of the experimental methods and results for 1974 and 1975 have been presented by Baker, Amacher, and Doty (*12*). Variable treatments included check plots with row or starter fertilizer only, plots receiving row fertilizer plus other commercial fertilizer in 1974 and 1975, plots treated with row fertilizer and dairy cattle manure at 5 dry tons per acre in 1974 and 1975, and plots treated with row fertilizer plus 5, 10, and 20 dry tons of a sewage sludge containing 257 ppm Cd on a dry matter basis. Since 1975, all plots have received only row fertilizer for the production of various food chain crops in order to study the residual effect of sewage sludge on crop yields and Cd accumulation in plant parts in relation to soil test levels of Cd.

It has been shown that chemical analysis of corn leaves removed from the node from which the ear emerges at the silking stage of maturity will reflect the soil availability of several elements (*13*). The results presented in Table 1 for corn leaves from plots receiving no sludge Cd are in close agreement with the range of 0.05 to 0.2 ppm reported as the normal range for plant leaves by Melsted in 1973 (*7*). His suggested maximum of 3 ppm Cd in plant leaves was reduced to 1 ppm Cd by Baker and Chesnin in 1975 (*2*) on the basis of the relationship between plant leaf Cd and the level expected in the food chain. While the upper limit of 1 ppm in corn leaves has no official validity, its use as a maximum level to be attained in corn ear leaves provides for interesting observations from the data summarized in Table 1. The lower values reported for plots receiving no sludge Cd in 1975 compared with those reported previously (*12*) resulted from an improved flameless atomic absorption technique which is capable of more accuracy when the level of Cd in plant tissue is below 0.1 ppm. The 5 dry ton rate of sludge supplied approximately 2.6 ppm Cd to the surface soil over the two-year period. The corn ear leaf Cd decreased from 1.09 ppm in 1974 after the first addition of 5 tons dry sludge to 0.4 ppm in 1976 and 1977. At all rates of sludge, the uptake of Cd from the second application was not greatly different from that observed for the initial application. This trend has been observed by other investigators and suggests that the availability is determined initially by the Cd binding properties of the sludge and not by the soil. However, over time as the sludge colloidal material is decomposed, the availability of Cd will be more a function of its chemistry in the soil. At the 10 and 20 dry ton rates of sludge the uptake of Cd was proportionately less than at the 5 ton rate. These results indicate that higher rates of Cd on fewer acres could result in less Cd entering the food chain. In the CAST Report No. 64 (*14*), it was reported that "repeated annual applications of sludge to soil cropped to corn show that the amounts applied in a given year influenced the Cd contents in leaves to a greater extent than did the total cumulative amounts of Cd applied." From our data as well as data reported by others (*14*, *15*), it is concluded that the plant accumulation of Cd from a given soil is a function of the total Cd loading of the soil, the properties of the sludge added, and the soil chemistry in relation to Cd binding once the sludge applications have been discontinued. However, once the soil abundance of Cd was increased to levels above the desired minimum, the plant concentration of Cd has attained levels in corn leaves above 1 ppm. Since it has not been possible to apply substantial quantities of Cd without increasing plant uptake of Cd, food chain buildup of Cd may be prevented only by preventing Cd additions to soil or by limiting the applications of Cd contaminated sludge to land dedicated to the production of non-food chain crops.

Table 1. Concentrations of Cd in ear leaf of corn plants at silking.[a]

	Cd, ppm			
	1974	1975	1976	1977
Check	0.08	0.03	0.06	0.06
Fertilizer	0.15	0.05	0.08	0.07
Manure—5[b]	0.15	0.07	0.07	0.09
Sludge—5	1.09	0.83	0.41	0.44
Sludge—10	1.66	0.95	0.87	1.06
Sludge—20	2.47	3.40	2.19	1.84

[a] A 1 ppm leaf concentration is currently recommended in Pennsylvania as a maximum.

[b] The numbers refer to tons of dry material per acre added to the field soil in 1974 and 1975.

Table 2. Soil test results for Cd obtained with three soil extractants in 1975 after two years of treatments with varying rates of Cd.

	Soil pH	Cd added, ppm[a]	Cd, ppm 0.1*N* HCl	DTPA[b]	Baker[c]
Check	6.7	—	0.12	0.08	0.06
Fertilizer	6.4	—	0.13	0.09	0.08
Manure—5	6.4	0.0013	0.11	0.08	0.06
Sludge—5	6.4	2.6	2.37	1.51	1.69
Sludge—10	6.5	5.2	5.62	3.18	2.66
Sludge—20	6.7	10.4	11.08	5.98	3.66

[a] Assumes an acre furrow slice to weigh 2,000,000 lb.
[b] Method of Lindsay and Norvell (*9*).
[c] Method of Baker (*12*).

Table 3. Soil test levels by Baker DTPA method.[a]

	Cd in soil, ppm			
	1974	1975	1976	1977
Check	0.09	0.06	0.09	0.15
Fertilizer	0.10	0.08	0.09	0.25
Manure—5	0.09	0.06	0.08	0.14
Sludge—5	2.35	1.69	1.19	2.02
Sludge—10	2.08	2.66	2.40	4.49
Sludge—20	8.48	3.66	5.89	8.17

[a] A test level of 1 ppm is recommended as the maximum level to be attained in soils of Pennsylvania.

Table 4. Soil test levels by Baker method converted to pCd which is analogous to soil pH.[a]

	pCd in soil test solution			
	1974	1975	1976	1977
Check	13.5	13.8	13.6	13.5
Fertilizer	12.9	13.6	13.4	13.0
Manure—5	13.2	13.8	13.5	13.4
Sludge—5	12.1	12.1	12.3	12.2
Sludge—10	11.9	11.9	12.0	11.8
Sludge—20	11.0	11.6	11.6	11.5

[a] A test level of 12.0 or greater is recommended for soils.

The amounts of Cd added by the various treatments and the quantities extracted by various testing methods may be compared from results presented in Table 2. Since the acre furrow slice treated with sludge was probably less than 2 million pounds, the results for 0.1*N* HCl suggest that this extractant removed all of the Cd added to the soil, while the Baker method, like the corn plants, removed proportionally less Cd as the soil level increased. The results by the Baker method for pH, Cd, Fe, Zn, Cu, Mn, and Ni are related to DTPA chemistry by a computer program to calculate the negative logarithm of the activity of each metal in each sample. The soil test results and the calculated pCd results are presented in Tables 3 and 4. From the results for the sludge-treated plots it does not appear that the test values are decreasing with time. When means for corn leaf Cd were correlated with soil test Cd and pCd over the four years, the respective coefficients were 0.82 and −0.84. Since the soils within the field were not extremely variable and the pH was not altered substantially by the treatments, a close correlation between soil test Cd and pCd ($r = -0.86$) was expected. However, where soil type and pH are expected to vary, the calculated pCd is used to obtain a theoretically superior measure of Cd availability. Both measurements are needed to predict the long term effect of sludge on Cd abundance as reflected by the amount removed with the testing solution and the current availability as reflected by pCd.

Table 5. Concentrations of Cd in cereal grains and potatoes grown on plots treated with different fertilizers and sewage sludge containing 257 ppm Cd on a dry weight basis.

	Cd, ppm Corn 1977	Corn 1975	Sorghum 1975	Wheat 1977	Potatoes 1977
Check	0.004	0.015	0.035	0.06	0.21
Fertilizer	0.004	0.003	0.108	0.10	0.28
Manure—5[a]	0.002	0.005	0.063	0.06	0.19
Sludge—5	0.024	0.025	0.470	0.60	0.88
Sludge—10	0.033	0.065	0.876	1.00	1.02
Sludge—20	0.048	0.120	0.855	1.42	1.45

[a] The numbers refer to the tons of dry material per acre added to the field soil in 1974 and 1975.

Table 6. Concentrations of Cd in liver, kidney, and muscle in meadow voles.[a]

Diets	Cd in diet, ppm	Cd in liver, ppm	Cd in kidney, ppm	Cd in muscle, ppm
Corn and synthetic (Syn) diets				
Syn	0.01	0.01[e]	0.03[f]	0.012[b]
Herbage (H)	0.10	0.08[e]	0.09[f]	0.018[b]
Syn + Cd	1.09	0.26[cde]	0.62[e]	0.023[b]
H + Cd	1.09	0.39[d]	0.62[e]	0.022[b]
Sludge H	1.09	0.43[c]	0.42[ef]	0.047[b]
Sorghum and synthetic (Syn) diets				
Syn	0.01	0.01[e]	0.03[f]	0.012[b]
Herbage (H)	0.23	0.03[e]	0.09[ef]	0.031[b]
Syn + Cd	2.76	0.63[c]	1.91[d]	0.027[b]
H + Cd	2.76	2.13[b]	3.69[b]	0.085[b]
Sludge H	2.76	1.86[b]	2.84[c]	0.025[b]

[a] Mean in column not followed by a common letter, differ significantly ($p < 0.05$). All results on a wet basis.

Table 7. Effect of dietary cadmium on the tissue cadmium concentration of liver, kidney, and muscle of young chicks.

Level of added dietary, Cd, μg/g	6 week body wt., g	Tissue cadmium content (after 6 weeks)[a] Liver, μg/g	Kidney, μg/g	Muscle, μg/g
0	615	0.23 ±0.04[b]	0.39 ± 0.01[b]	0.07 ± 0.01[b]
3	655	4.75 ± 1.98[b]	9.27 ± 2.25[c]	0.15 ± 0.02[b]

[a] Means ± standard deviation dry fat-free basis. Means not followed by the same letter are significantly different ($p < 0.05$).

Table 8. Effect of dietary cadmium on egg production, shell thickness, and tissue cadmium concentration in the laying hen (12 month experiment)

Level of added dietary Cd, μg/g	Production, %	Thickness, mg/cm²	Tissue Cd content[a] Egg powder, μg/g	Muscle, μg/g	liver, μg/g	Kidney, μg/g
0	60.4[b]	77.2[b]	0.16 ± 0.05[b]	0.12 ± 0.04[b]	2.99 ± 1.3[b]	17.1 ± 13.4[b]
3	68.9[b]	77.8[b]	0.13 ± 0.04[b]	0.57 ± 0.36[b]	33.47 ± 18.4[b]	273.9 ± 45.8[c]

[a] Means ± standard deviation dry fat-free basis. Means not followed by the same letter are significantly different ($p < 0.05$).

Accumulation of Cd in the Food Chain

The concentrations of Cd in cereal grains and potatoes for different field treatments are presented in Table 5. Except for corn grain, the crops accumulated substantial quantities of Cd. Two years after the last application of sludge the corn grain contained about 10 times the background level for check plots; the wheat contained about 15 times the background levels; and potatoes contained 10 to 12 times the background levels. Unless sludge containing Cd is used exclusively for the production of corn for grain which is relatively low in Cd, the effect on the food chain can be expected to be about equal for all these crops.

Cadmium in Animal Tissues

In order to assess the impact of increased plant Cd content on the food chain, several animal feeding trials have been conducted for the purpose of determining the relationship between dietary and tissue Cd content. In one study, meadow voles (*Microtus Pennsylvanias*) were fed herbage grown on sludge fertilized plots (*16*). Diets containing the sludge fertilized corn herbage had a Cd content of 1.09 ppm while the diets containing sludge fertilized sorghum contained 2.76 ppm Cd. For the purpose of determining the relative availability of Cd, the synthetic control diet and diets containing herbage grown on check plots were supplemented with Cd salts to give comparable levels of dietary Cd. The results are presented in Table 6. All diets containing supplemental Cd resulted in significant increases in liver and kidney Cd content. Muscle showed a greatly reduced ability to accumulate Cd. Although cadmium supplementation resulted in an increase in muscle Cd content, these changes did not prove to be statistically significant. The availability of Cd in herbage from sludge amended plots was equal to or greater than Cd added as soluble Cd to the herbage from check plots.

Feeding trials have been conducted with broiler chicks and laying hens (*17*). Normal diets which contained 0.20 ppm Cd were supplemented with 3 ppm Cd in order to approximate the levels observed with sludge treated herbage. The data from these experiments are presented in Tables 7 and 8. The results were very similar to those obtained with the meadow voles. Cadmium readily accumulated in liver and kidney while muscle was not as sensitive to dietary Cd content. Although the results are not statistically significant, 3 ppm dietary Cd consistently doubled muscle cadmium content. There was no transfer of cadmium to the egg in this experiment. These results are consistent with observations of Sell (*18*), who studied the transfer of Cd^{109} to the egg, as well as the results of Mills and Delgarno (*19*), who reported poor placental transfer of cadmium in sheep.

The results of these studies show that the amounts of Cd found in plants grown on sludge amended soils can increase the Cd content of tissues such as liver and kidney. Although there was not a statistically significant increase in muscle Cd content, these amounts of dietary Cd (1–3 ppm) consistently doubled muscle cadmium content. Interpretation of these results in relation to human intake of cadmium and long range health effects of Cd is required for the proper monitoring of sewage sludge application on land used for production of crops which enter the food chain.

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