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Accumulation of heavy metals in plants and potential phytoremediation of lead by potato, *Solanum tuberosum* L.

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The use of sewage sludge as a source of nutrients in crop production is increasing in the United States and worldwide. A field study was conducted on a 10% slope at Kentucky State University Research Farm. Eighteen plots of 22 × 3.7 m each were separated using metal borders and the soil in six plots was mixed with sewage sludge, six plots were mixed with yard waste compost, and six unamended plots were used for comparison purposes. During a subsequent 3-year study, plots were planted with potato (year 1), pepper (year 2), and broccoli (year 3). The objectives of this investigation were to: (i) characterize chemical properties of soil-incorporated sewage sludge and yard waste compost; (ii) determine the concentration of seven heavy metals (Cd, Cr, Ni, Pb, Zn, Cu, and Mo) in sewage sludge and yard waste compost used for land farming; and (iii) monitor heavy metal concentrations in edible portions of plants at harvest. Concentrations of heavy metals in sewage sludge were below the U.S. EPA limits. Analysis of potato tubers, peppers, and broccoli grown in sludge-amended soil showed that Cd, Cr, Ni, and Pb were not significantly different from control plants. Concentrations of Zn, Cu, and Mo were significantly greater in tubers and peppers grown in sludge compared to their respective controls. Zn and Mo in broccoli heads were higher than their control plants. The ability of potato to accumulate lead needs additional investigation to optimize the phytoremediation of this pollutant element.

Keywords: Biosolids, yard waste compost, soil conditioners, pepper fruits, broccoli heads.

Introduction

The increased production of sewage sludge in the United States has led many municipalities to consider the application of sewage sludge to agricultural land as a means of sludge and nutrient recycling. The U.S. Environmental Protection Agency (U.S. EPA) promotes beneficial use of municipal solids because it decreases dependence on chemical fertilizers and provides significant economic advantages. Sewage sludge (biosolids) contains organic matter, and macro and micronutrients important for plant growth. Sixteen elements out of the 90 found in plants, known to be essential for plant growth, are present in biosolids. Some of these elements, however, can be detrimental to human, plant or animal life if they are present above certain limits. These detrimental elements are regulated by respective statute.

The value of biosolids is their ability to improve the soil by providing plant nutrients and by improving soil structure and other characteristics. The potential value of these to the farmlands cannot be questioned. Efficient and effective use of these materials as soil conditioners provides one of the best means we have for maintaining and restoring soil productivity. Biosolids from different origins have unique properties that should be thoroughly investigated in the soil/water/plant ecosystem. In addition, the simultaneous use of soil conditioners to enhance soil physical, chemical, and microbial conditions could also enhance soil bioremediation.

The sharply escalating production costs associated with the increasing costs of energy and fertilizers to U.S. farmers and the problems of soil deterioration and erosion associated with intensive farming system have generated considerable interest in less expensive and more environmentally compatible production alternatives such as recycling wastes from several processing operations to produce high-quality organic amendments for soil improvement and crop production. On the other hand, accumulation of heavy metals by plants grown on sewage sludge-treated soils[1,2] requires a better understanding. Elevated Cd concentrations in soil, resulting from the application of biosolids has been perceived as a potential environmental hazard.[3,4]

The primary Cd risk posed by the agricultural use of biosolids is the increased dietary Cd intake of people consuming crops grown on these soils. There is limited

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information on heavy-metal absorption by vegetable crops from sludge-treated soil. The extent of heavy metal accumulation in plant tissue appears to be affected by many soil-, plant-, and sludge-related factors. The rate of release of heavy metals in sewage sludge into soil solution and subsequent uptake by plants could result in phytotoxicity. Elevated concentrations of heavy metals in harvested plant tissue could expose consumers to excessive levels of potentially hazardous chemicals. Identifying management strategies that meet crop nutrition needs, support crop production, and protect food quality is the focus of this investigation.

The objectives of this investigation were to (1) characterize chemical properties of soil incorporated sewage sludge and yard waste compost; (2) determine the concentration of seven heavy metals (Cd, Cr, Ni, Pb, Zn, Cu, and Mo) in sewage sludge and yard waste compost; and (3) monitor heavy metal concentration in edible portions of plants at harvest.

Materials and methods

Field study

A field study was conducted on a Lowell silty loam soil (2.8% organic matter, pH 6.9) located at Kentucky State University Research Farm, Franklin County, KY. The soil has an average of 12% clay, 75% silt, and 13% sand. Eighteen (18) universal soil loss equation (USLE) standard plots of 22×3.7 m each were established on a soil of 10% slope. Plots were separated using metal borders 20 cm above ground level to prevent cross-contamination between adjacent treatments. Three soil management practices were used: (1) sewage sludge (obtained from Nicholasville Wastewater Treatment Plant, Versailles, KY) was mixed with native soil at 30 t acre^{-1} (on dry weight basis) with a plowing depth of 15 cm, (2) yard waste compost made from yard and lawn trimmings, and vegetable remains (obtained from Kentucky State University Research Farm, Franklin County) was mixed with native soil at 30 t acre^{-1} (on dry weight basis) with a plowing depth of 15 cm, and (3) a no-mulch (NM) control treatment (roto-tilled bare soil) was used for comparison purposes.

In year 1, potato (*Solanum tuberosum* cv. Kennebec) seed pieces were planted in 10 rows plot^{-1} (10 plants row^{-1}). Plots were irrigated by drip tape (Rainbird Corporation, Glendora, CA) and no fertilizer was applied. Trifluralin (Treflan; 430 g liter^{-1} EC) was sprayed on the soil surface at the rate of 0.75 lb acre^{-1} and incorporated into the soil. In year 2, sweet pepper (*Capsicum annuum* L. cv. Aristotle-X3R) seedlings 60 days old, were planted at 10 rows plot^{-1} along the contour of the land slope at 10 plants row^{-1}and napropamide (Devrinol 50-DF; 4 lbs formulated product acre^{-1}) was sprayed on soil as a pre-emergent herbicide. In year 3, broccoli (*Brassica oleracea* L. cv. Packman F1) seedlings 45 days old, were planted at 10 rows plot^{-1} along the contour of the land slope at 10 plants row^{-1} and napropamide was used.

Soil and plant tissue analysis

Soil and soil-incorporated sewage sludge and yard waste samples were collected to a depth of 15 cm from field plots using a soil core sampler equipped with a plastic liner (Clements Associates, Newton, IA, USA) of 2.5 cm i.d. Soil samples were oven-dried at 65°C for 48 hours and then sieved to a size of 2 mm. pH was determined using a glass electrode in a soil: distilled water slurry (1:5, w/v). Soil organic matter was calculated as dry weight minus ash content. Nitrogen was determined by the Kjeldahl method. Quantitative analyses of Mehlich-3 extractable Cd, Cr, Ni, Pb, Zn, Cu, and Mo were conducted using an inductively coupled plasma[5] (ICP, Varian Vista-Pro) spectrometer. Detection limits (mg/kg) were Cd 0.02, Cr 0.04, Cu 0.04, Mo 0.1, Ni 0.2, Pb 0.3, Zn 0.04 at wavelengths (nm) 226.502, 267.716, 324.754, 202.032, 231.604, 220.353, and 213.857, respectively.

At harvest 25 potato tubers, pepper fruit or broccoli heads of comparable size were collected at random from each of the 18 field plots (six replicates for each soil treatment), washed with tap and deionized water and dried in an oven at 65°C for 48 hours. The dried samples were ground manually with ceramic mortar and pestle to pass through 1 mm sieve. Samples were re-dried to constant weight using an oven. To 1 g of each dry sample, 10 mL of concentrated nitric acid was added and the mixture was allowed to stand overnight, and then heated for 4 hour at 125°C on a hot plate. The mixture was then diluted to 50 mL with double distilled water and filtered through filter paper No.1. Concentrations of Cd, Cr, Ni, Pb, Zn, Cu, and Mo were determined using ICP spectrometry.

Results and discussion

Sewage sludge application altered the chemical and physical properties of soil, which in turn affected soil nurient balance (Table 1). Addition of sludge also increased the soil pH about 1.5 units compared to native soil. Soil pH affects ion availability.[6]An increase in pH can bring about strong adsorption on soil particles or in some cases, precipitation of Cu and Zn among other metals, which in turn allows for lower accumulation of these metals in plant tissues.[7]

Sewage sludge contains great amounts of nutrients especially N, P, and Ca (Table 1) that plants require. P concentrations in sewage sludge reached levels comparable with super-phosphate fertilizer.

As expected, total N and C were greater in the 1–15 cm soil horizon as a result of the addition of sewage sludge. Total C was 3.8 vs. 1.6% and total N was 0.4 vs. 0.1% in the unamended vs. sludge-amended soils, respectively.

Table 1. Chemical characteristics of sewage sludge, yard waste compost, and soil incorporated with sewage sludge or yard at Kentucky State University Research Farm, Franklin County, Frankfort, Kentucky, USA, 2001

Soil parameters	*Sewage sludge*	*Yard waste compost*	*Native soil*	*Sewage sludge incorporated with native soil*	*Yard waste incorporated with native soil*
pH*	12.33 ± 0.77 a	7.05 ± 0.17 c	6.99 ± 0.02 c	8.48 ± 0.13 b	7.31 ± 0.22 c
Organic Matter** %	30.26 ± 1.15 a	21.07 ± 0.16 b	2.72 ± 0.51 d	7.27 ± 0.42 c	7.60 ± 0.33 c
C %	17.05 ± 0.56 a	13.02 ± 1.36 b	1.59 ± 0.06 d	3.77 ± 0.35 c	3.84 ± 1.03 c
N %	2.16 ± 0.13 a	1.02 ± 0.07 b	0.15 ± 0.01 d	0.39 ± 0.02 c	0.32 ± 0.07 c
C/N ratio	7.90 ± 0.24 d	12.74 ± 0.44 a	10.54 ± 0.53 b	9.54 ± 0.47 c	11.78 ± 0.68 a
P %	1.23 ± 0.32 a	0.66 ± 0.02 b	0.18 ± 0.06 c	0.31 ± 0.13 c	0.24 ± 0.12 c
K %	0.20 ± 0.01 b	0.67 ± 0.16 a	0.25 ± 0.07 b	0.23 ± 0.06 b	0.28 ± 0.10 b
Ca %	19.10 ± 1.28 a	5.18 ± 0.10 b	0.40 ± 0.15 d	2.85 ± 1.19 c	1.04 ± 0.54 d
Mg %	0.38 ± 0.08 b	0.57 ± 0.03 a	0.19 ± 0.08 c	0.22 ± 0.11 bc	0.25 ± 0.09 bc
Cd ppm	0.13 ± 0.04 a	0.30 ± 0.09 a	0.25 ± 0.06 a	0.25 ± 0.05 a	0.18 ± 0.03 a
Cr ppm	0.40 ± 0.12 a	0.08 ± 0.02 c	0.15 ± 0.03 bc	0.30 ± 0.08 ab	0.13 ± 0.02 c
Ni ppm	6.0 ± 0.8 a	0.28 ± 0.06 c	0.63 ± 0.05 c	1.73 ± 0.06 b	0.85 ± 0.14 c
Pb ppm	0.50 ± 0.10 b	4.65 ± 0.92 a	0.85 ± 0.12 b	0.70 ± 0.09 b	1.53 ± 0.22 b
Zn ppm	66.93 ± 5.22 a	24.18 ± 2.74 b	3.95 ± 0.98 e	20.13 ± 2.50 c	7.78 ± 1.25 d
Cu ppm	94.75 ± 7.85 a	2.25 ± 0.55 c	1.28 ± 0.42 c	17.23 ± 2.66 b	1.98 ± 0.75 c
Mo ppm	0.63 ± 0.05 a	0.01 ± 0.00 b	0.08 ± 0.02 b	0.09 ± 0.01 b	0.06 ± 0.03 b

Each value in the table is an average obtained from analysis of six samples. Values within a row followed by the same letter(s) are not significantly different ($P < 0.05$) (SAS Institute, 2001; Duncan's multiple range test).[15] *pH was determined using a glass electrode in a soil: distilled water slurry (1:5, w/v). **Soil organic matter was calculated as dry weight minus ash content. †Sewage sludge and yard waste compost were each mixed with native soil at 30t acre^{-1} on dry weight basis.‡ Nitrogen was determined by Kjeldahl method. All other elements were determined using an Inductively Coupled Plasma Spectrometer.

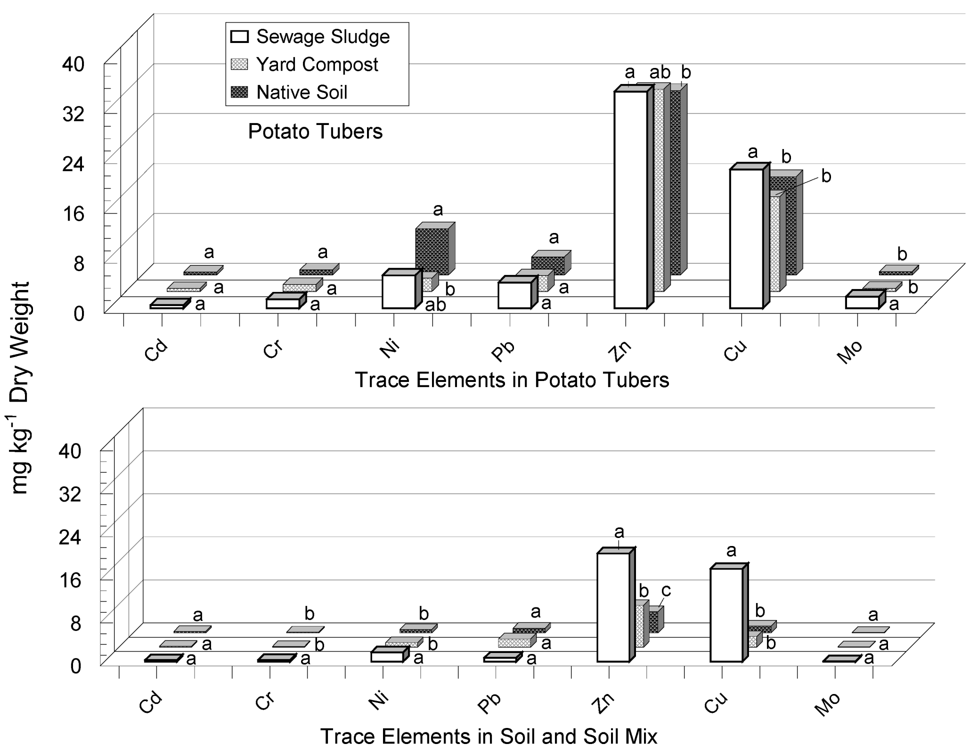

Fig. 1. Mean concentrations of Cd, Cr, Ni, Pb, Zn, Cu, and Mo in tubers of potato plants grown under three management systems (upper graph) at Kentucky State University Research Farm and concentrations of heavy metals in soil amended with sewage sludge or yard waste compost compared to native soil (lower graph). Statistical comparisons ($P < 0.05$) were carried out between three soil management practices for each element. Bars for an element accompanied by the same letter(s) are not significantly different using the ANOVA procedure.[15]

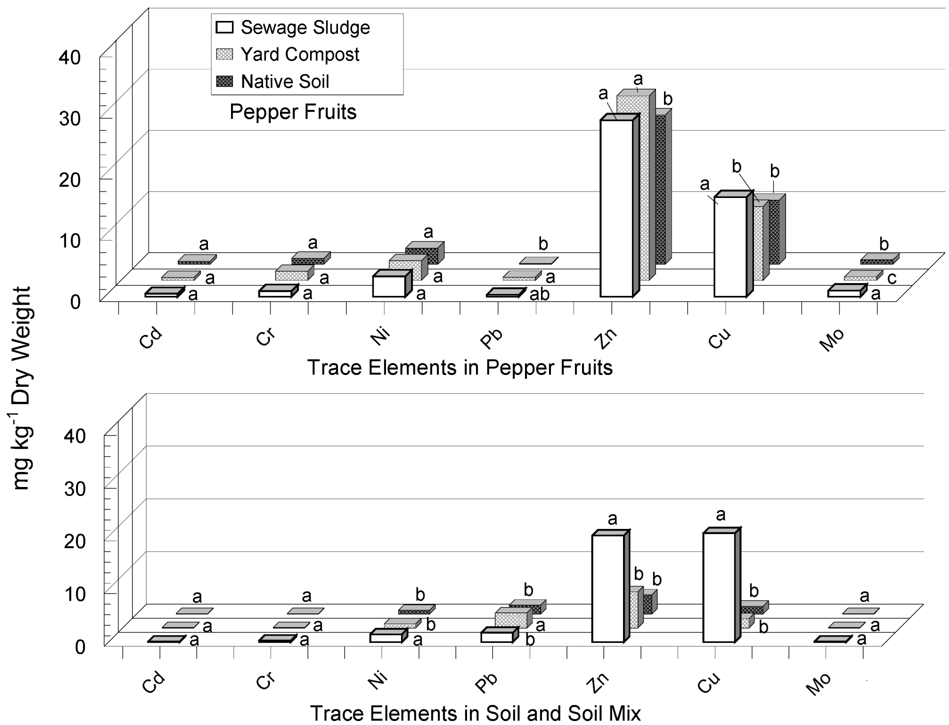

Fig. 2. Mean concentrations of Cd, Cr, Ni, Pb, Zn, Cu, and Mo in fruits of pepper plants grown under three management systems (upper graph) at Kentucky State University Research Farm and concentrations of heavy metals in soil amended with sewage sludge or yard waste compost compared to native soil (lower graph). Statistical comparisons ($P < 0.05$) were carried out between three soil management practices for each element. Bars for an element accompanied by the same letter(s) are not significantly different using the ANOVA procedure.[15]

However, sludge can also contain toxic metals, although at what level and when such metals might cause harmful effects are largely unknown.[8] The U.S.EPA has defined clean sludge in terms of its heavy metal content (mg kg^{-1}; Zn 1400, Cu 1500; Ni 420, Cd 39; Pb 300; Cr 1200; Mo 75). Unlimited amounts of sludge could be added to land if all these metals were below their limit.

Generally, the concentrations of heavy metals in sewage sludge used in this study were below the allowable limits and therefore this sludge has potential for agricultural use. However, results in Figure 1 (upper graph) indicated that Zn and Cu were accumulated in potato tubers grown even in the no-mulch soil. These data are consistent with Morrison et al.[9] who found that plants rapidly accumulate Cu. Soil analysis during the 3 years of the study revealed that Zn and Cu have increased significantly in soil as a result of sludge addition.

Plant uptake is one of the main pathways through which metals enter a food chain. This pathway transfers the metals through higher trophic levels to humans. Although Zn has relatively low toxicity to humans, studies have shown allergies and zinc poisoning could occur along the food chain, which may also interfere with copper metabolism.[10] Zn and Cu concentrations in sewage sludge obtained from the Nicholasville Wastewater Plant were extremely high (66.9 and 94.7 mg kg^{-1}, respectively) compared to other trace metals in sewage sludge. However, these concentrations are below the pollutant concentration limit in sewage sludge.[8] Zn and Cu concentrations were significantly reduced to 20.1 and 17.2 mg kg^{-1}, respectively when sludge was incorporated with native soil (Table 1). Generally, Zn and Cu levels in sludge and plants were of no major concern in the present study.

Cd and Pb are the heavy metals of greatest concern to human health since plants can take them up and introduce them into the human food chain. Levels of Cd and Pb in soil amended with sewage sludge averaged 0.25 and 0.7 mg kg^{-1}, respectively. These levels are much lower than the limits in the U.S. guidelines for using sewage sludge in land farming. Thus there was no major concern posed by Cd and Pb levels in sewage sludge for use as agricultural fertilizer. Our results revealed that concentrations of Cd in potato tubers, pepper fruits, and broccoli heads were 0.50, 0.49, and 0.04 mg kg^{-1}, respectively (Figs.1–3). Data for all crops analyzed in this investigation are expressed on dry weight basis. Considering that water content of potato was 85% and that of pepper was 95%, the Cd concentrations were near their Codex-established maximum limit[11] of 0.1 mg kg^{-1} for potato and 0.05 mg kg^{-1} for pepper.

Pb is defined by USEPA as potentially toxic to most forms of life. According to Codex Standard 230-2001, Revision 1-2003,[12] the maximum level for lead in most vegetables, including peeled potatoes, is 0.1 mg kg^{-1}. For

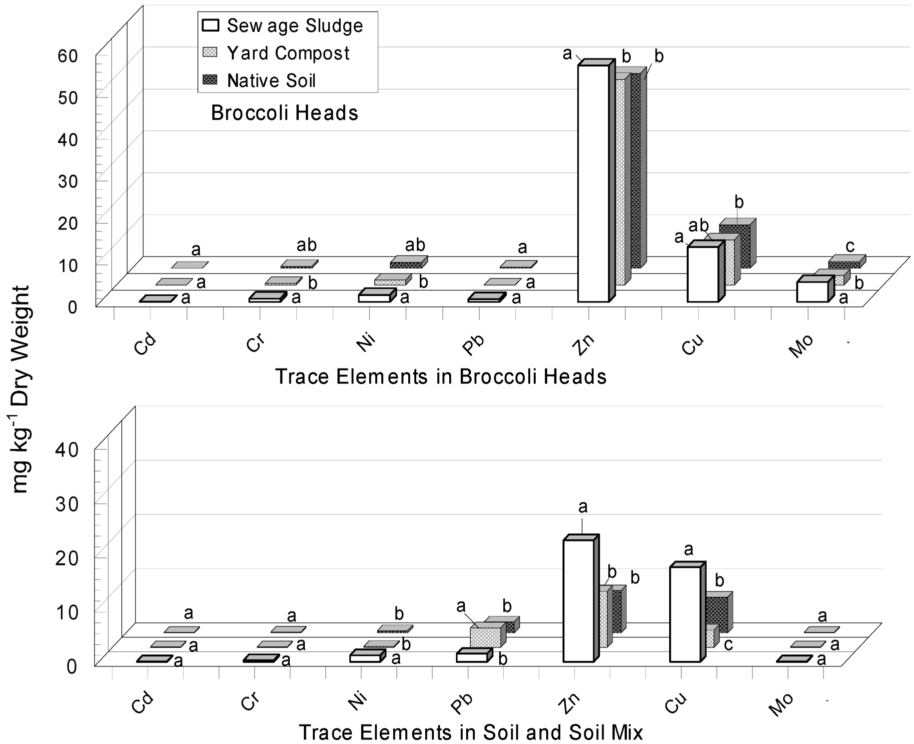

Fig. 3. Mean concentrations of Cd, Cr, Ni, Pb, Zn, Cu, and Mo in heads of broccoli plants grown under three management systems (upper graph) at Kentucky State University Research Farm and concentrations of heavy metals in soil amended with sewage sludge or yard waste compost compared to native soil (lower graph). Statistical comparisons ($P < 0.05$) were carried out between three soil management practices for each element. Bars for an element accompanied by the same letter(s) are not significantly different using the ANOVA procedure.[15]

cruciferous vegetables such as broccoli, the maximum level is 0.3 mg kg^{-1}. Based on water content calculations obtained for potatoes and peppers, the maximum lead levels were exceeded regardless of soil composition in potatoes, but not in peppers. However, the potatoes analyzed for this study were not peeled, and peeled potatoes may have reduced lead levels.

Accumulation of the seven heavy metals varied between plant species. The different absorption pattern of heavy metals among the three vegetables (potato, peppers, and broccoli) investigated in this study could be attributed to individual plant characteristics. There was a significant increase in concentrations of Zn and Cu in potato tubers grown under the three soil management practices investigated in this study even in potato grown in native soil. A comparison of heavy metal accumulation among the three vegetable species was not an objective of the study. Different crops were grown in different years, which makes valid statistically comparisons difficult. With these cautionary statements in mind, a post-hoc analysis indicated that Pb concentrations were significantly higher in potatoes 3.19 $\pm$ 0.20, compared to pepper 0.31 $\pm$ 0.14 and broccoli 0.33 $\pm$ 0.16 mg kg^{-1} dry wt. It is possible that potato will accumulate Pb, and could serve as a species useful for bioremediation. This needs additional investigation.

There is an urgent need to develop long-term, low-energy, biological, self-sustainable systems of farming. Recycling wastes from several processing operations for production of high quality organic amendments is simple, inexpensive, energy conserving, and effective for erosion control (data not shown) and nutrient recycling. Our previous studies have also indicated that the use of sewage sludge in land farming can become a useful technique for trapping pesticides such as trifluralin[13]and may reduce surface and groundwater contamination by other commonly used pesticides. On the other hand, research has indicated that increased dissolved organic carbon (DOC) in sewage sludge decreases the adsorption of metals to soil surfaces through the formation of organometalic complexes,[14] thereby increasing the bioavailability of metals to plants.

The impact of potentially toxic trace metals in sludge applied to cropland can be reduced by growing crops that do not accumulate these metals in their edible portions, or by increasing soil pH to bring about strong adsorption of metals on soil particles,[6,7] and by reducing the rate at which sludge is applied. Future research should consider variations in uptake between different plant species, the level of trace metals present in the atmosphere surrounding the study area, and explore plant genetic resources for nutritional improvement and phytoremediation. Additionally, the long-term effect of sewage sludge compost on the accumulation of heavy metals in edible plants should also be investigated to prevent elevated concentrations of heavy metals from reaching the consumer.

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References

[1] Brallier, S.; Harrison, R.B.; Henry, C.L.; Dongsen, X. Liming effects on availability of Cd, Cu, Ni, and Zn in a soil amended with sewage sludge. Water Air Soil Poll **1996**, *86*, 195–206.
[2] McGrath, S.P.; Zhao, F.J.; Dunham, S.L.; Crosland, A.R.; Coleman, K. Long-term changes in the extractability and bioavailability of Zinc and Cadmium after sludge application. J. Environ. Qual. **2000**, *29*, 875–883.
[3] Alloway, B.J.; Jackson, A.P. The behavior of heavy metals in sewage-sludge amended soils. Sci. Total Environ. **1991**, *100*, 151–176.
[4] McBride, M.B. Toxicity of metal accumulation from agricultural use of sludge. J. Environ. Qual. **1995**, *24*, 5–18.
[5] Vazquez, E.; Cancela, R.; Castro, M.; Gonzalez, A.; Abreu, C.A. Trace elements extracted by DTPA and Mehlich-3 from agricultural soils with and without compost additions. Commun. Soil Sci. Plant Anal. **2005**, *36*, 717–727.
[6] Woodbury, P.B. Trace elements in municipal solid waste composts. Biomass Bioene **1992**, *3*, 239.
[7] Stratton, M.L.; Rechcigl, J.E. Organic mulches, wood products, and compost as soil amendments and conditioners. In: *Handbook of Soil Conditioners*. Wallace A.; Terry R Eds, Marcel Dekker, Inc., New York, 1998, 43–95.
[8] U.S.EPA. *Standards for Disposal of Sewage Sludge; Final Rules*: Part II. Federal Register, February 19, 1993; pp. 9247–9415.
[9] Morrison, G.; Fatoki, O.S.; Linder, S.; Lundehn, C. Determination of heavy metal concentrations and metal fingerprinting of sewage sludge. Water, Air Soil Poll **2004**, *152*, 111–127.
[10] Ohnessorge, F.K.; Wilhelm, M. Zinc. In: *Metals and their Compounds in the Environment: Occurrence, Analysis and Biological Relevance*, Merian, E., Ed., VCH Weinheim, New York 1991, 1330.
[11] Codex Maximum Levels 2005. Cadmium: http://www. codexalimentarius.net/ download/standards/10243/CXS_248e.pdf
[12] Codex Maximum Levels 2003. Lead: http://www. codexalimentarius.net/download/ standards/372/CXS_230e.pdf
[13] Antonious, G.F.; Patterson, M.A.; Snyder, J.C. Pesticide residues in soil and quality of potato grown with sewage sludge. Bull. Environ. Contam. Toxicol. **2003**, *71*, 315–322.
[14] Fjallborg, B.; Dave, G. Toxicity of Sb and Cu in sewage sludge to terrestrial plants and sewage elutriate to aquatic organisms and its interaction. Water, Air Soil Poll **2004**, *155*, 3–20.
[15] SAS Institute, *SAS/STAT Guide*, Release 0.03 Edition, SAS Inc., SAS Campus Drive, Cary, NC 27513, USA, 2001.