Compost Science & Utilization, (2005), Vol. 13, No. 1, 60-64

Heavy Metal Accumulation in a Sandy Soil and in Pepper Fruit Following Long-term Application Of Organic Amendments

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Heavy metals are toxic and persistent pollutants that may be present in organic waste materials used as soil amendments. Following accumulation in amended soil, crop plants could assimilate these pollutants in sufficient concentrations to pose a threat to humans consumers. The Federal Clean Water Act and additional state regulations set minimum standards for heavy metals in organic amendments. Cumulative amounts of heavy metal pollutants that may be added to soils through amendments are regulated through national and state environmental protection agencies and the Federal Food and Drug Administration regulate maximum tolerances of heavy metal contaminants in food. The objective of the research was to study the effects of long-term organic amendment application on the accumulation of heavy metal pollutants in soil and subsequent contamination in pepper fruits. Organic amendments were applied yearly to replicate large plots during 1996 to 2000. Controls received no amendments. Different organic amendments were applied every year to simulate grower organic amendment availability throughout long-term application. Although higher levels of extractable Cd, Cu, Ni, Pb, and Zn were observed from amended soils during 1996 to 2000, all were within acceptable levels. Furthermore, no accumulation of Cd, Cu, Pb, and Ni was observed in pepper fruit. Therefore, long-term application of organic amendments made from waste materials with pollutant content below maximum acceptable levels under state and federal regulations should be suitable for vegetable production.

Introduction

The benefits of organic amendments to growth and yield of vegetables and other crops on Florida's sandy soils have been clearly demonstrated (Ozores-Hampton, *et al.* 1998, Ozores-Hampton and Peach 2002). Such recycling also provides obvious benefits to municipalities seeking to dispose of what would otherwise be organic wastes. Nevertheless, concerns over possible contamination still exist among consumers when vegetables are grown in soil that have been amended with MSW (municipal solid waste), biosolids (BS), food processing residuals, manures, yard trimming waste (YTW), and agricultural by-products, despite 30 year of research in the area (Chaney 1994). Toxic metals may accumulate in the soil (Sterrett *et al.* 1982; Yuran and Harrison 1986) or to be taken up and accumulated in the edible plant parts, where they pose a potential threat to consumers (Shiralipour *et al.* 1992). Metals posing the greatest threat to human health are Cd, Cu, Pb, Ni, and Zn (Chaney 1993).

The Florida Department of Environmental (FDEP) currently regulates allowable concentrations of five pollutant metals in compost made from waste materials (FDEP 1989). Maximum permissible concentrations of Cd, Cu, Pb, Ni, and Zn for code 1 MSW are 15, 450, 500, 50, and 900 $mg \cdot kg^{-1}$ dry weight respectively (FDEP 1989). The state also limits the total cumulative amount of Cd, Cu, Pb, Ni, and Zn applied in MSW compost to the soil to 5, 124, 499, 124, and 249 $kg \cdot ha^{-1}$ respectively (DEP 1989). Presently, biosolids and biosolids mixed with either YTW or MSW are regulated at the Federal level under the Clean Water Act Section 503 (USEPA 1994, 1995). Clean Water Act Section 503 classified biosolids quality with respect to the nine regulated pollutant elements concentration limit as the pollutant ceiling concentration and pollutant concentration and two loading rates based limit, cumulative pollutant loading rates (CPLR) and annual pollutant loading rates [(APLR) USEPA 1994 1995]. Eighteen states have regulations in place that are more restrictive than Section 503 (Goldstein 2000). Evaluation of potential food chain transfer of Cd, Cu, Pb, Ni, and Zn in compost shows that consumption for 70 years of 60% of garden food produced on pH 5.5 soils amended with 1,000 $Mg.ha^{-1}$ of compost would be safe (Chaney 1994). Biosolids or compost made from waste materials that do not meet EPA 503 standards for pollutant concentration should not be applied to horticultural land (Ozores-Hampton and Peach 2002). However, few field studies have investigated the long-term cumulative effects of organic amendments application in vegetable production (Ozores-Hampton *et al.* 1994a and b; Ozores-Hampton *et al.* 1997). The objective of this research was to study the effects of long-term organic amendments application on the accumulation of heavy metals in soil and peppers fruits.

Materials and Methods

Experiments were conducted during 1993 to 2000 at the University of Florida's Southwest Florida Research and Education Center in Immokalee. The Soil and Water Science Department, Univ. of Florida, Gainesville analyzed chemical and physical properties of the organic amendments.

Field Procedures

Soil was an Immokalee fine sand (sandy, siliceous, hyperthermic Arenic Haplaquods). Treatments consisted of a yearly organic amendment application or nonamended controls plots. Different organic amendments were applied every year to simulate grower organic amendment availability throughout long-term application (Table 1). The experiment used is a randomized block design with four replications of each treatment.

Cucumber (*Cucumis sativus* L.), broccoli (*Brassica oleracea* Italica L.), eggplant (*Solanum melongena* L.), squash (*Cucurbita pepo* L.), tomatoes (*Lycopersicon esculentum* Mill.), and watermelons (*Citrullus vulgaris* Schrad.), were grown during 1993 to 1998 spring seasons. A rotation of peppers (*Capsicum annuum* L.) and watermelon were grown from 1998 to 2000. Organic amendments were applied at bed formation in the fall each year. Methyl bromide and chloropicrin (98:2) were used to fumigate the soil and applied at bed formation at the rate of 336 $kg.ha^{-1}$ each year. Beds were covered with white-faced black polyethylene mulch. Drip irrigation tubing provided with emitters every 0.15 $m^2 \cdot h^{-1}$ was positioned in the center of the bed prior to plastic mulch application. Pepper crops were transplanted in the fall onto raised bed 0.81 m wide, 0.1 m high, and 1.8 m between centers in 85 m long plots. Spacing was 25 cm in double rows separated by 45 cm giving plant populations of 43,243 $plants \cdot ha^{-1}$. Peppers were harvested two to three times during the season. Watermelons were planted into the beds where the pepper crop had grown. Spacing was 1.8 m between plants giving a 3,136 $plants.ha^{-1}$. Watermelons were harvested an average of three times during the seasons.

Fertigation rates of nitrogen were reduced by 50% in amended plots beginning in 1998 to compensate for mineralization from the organic amendments. Fertilizer was applied to the vegetables by injection through the drip irrigation system following the Univ. of Florida Extension guidelines (Hochmunth and Maynard 1998). Plants were monitored for insects and diseases and pesticides were applied, as needed according to Univ. of Florida Extension guidelines (Hochmunth and Maynard 1998).

TABLE 1.
History of organic amendments applied to the soil during 1993 to 2000 seasons

Year	Organic Amendments	Rate ($Mg \cdot ha^{-1}$)	Source (Florida)
1993	Municipal solid waste compost	180	Broward County
1994	Biosolids	8	Tampa
1995	Yard trimmings and biosolids compost	23	Palm Beach
1996	Yard trimmings and biosolids compost	45	Palm Beach
	Cow manure compost	27	Oxford
1997	Yard trimmings and biosolids compost	45	Palm Beach
1998	Biosolids (Class B)	38	Miami
1999	Biosolids (Class B)	47	Miami
2000	Biosolids (Class B)	38	Miami

Chemical Analysis

Moisture concentration was obtained by oven drying 10 g (wet weight) of organic amendment at 105°C for 24 h. Total N and C concentrations were measured in organic amendment samples that were air dried for 4 days, ground in a Spex 8000 Mixer/Mill, and combusted at 1010°C in a Carlo-Erba NA-1500 C/N/S analyzer. The organic amendment samples were acid digested and analyzed by Inductively Coupled Argon Plasma Spectroscopy (ICAP). Total nutrients and trace metals were analyzed according to EPA Method 3050 (USEPA 1990).

Soil samples were collected (500 g) from the center of the bed at the end of the season during 1996-2000. Samples were oven dried at 28°C and extracted with nitric acid. Cadmium, Cu, Pb, Ni, and ZN were determined by Inductively Coupled Argon Plasma Spectroscopy (ICAP) according to EPA Method 3050 (USEPA 1990). Five fruits were randomly collected from pepper plants immediately before the first harvest from organic and nonorganic amended plots during 1998 and 1999 season. Fruit samples were washed in tap water, dried at 60°C, ground in a stainless steel mill, and analyzed for Cd, Cu, Pb, Ni, and

Zn concentrations according to AOAC Method 975.03 (AOAC 1996)

Soil accumulative pollutant loading rate was obtained by multiplying the soil pollutant concentration by the weight of a hectare of soil, taken to be 10^6 kg. Data were subjected to analysis of variance (ANOVA) and mean separation according to Duncan's Multiple Range Test.

Results and Discussion

Organic Amendments

Organic amendments showed a near neutral to alkaline pH, a C:N ratio below 20, from 1.0 to 6% N and 0.3 to 3% phosphate (P_2O_5, Table 2). Pollutant concentrations of the organic amendments were below maximum acceptable levels under Florida Department of Environmental Protection (FDEP 1989) and Federal level under Clean Water Act Section 503 (USEPA 1994 1995).

Soil Concentrations And Loading Rates

Significantly higher levels of extractable Cd, Cu, Ni, Pb, and Zn were observed in organic amendments compared to nonamended soil (Table 3). However total soil accumulative pollutant loading rates by year 2000 were still orders of magnitude below minimum requirements of both state and federal regulations (Table 4). Cadmium, Cu, Pb, Ni, and Zn levels in the organic amendment plots were within normal ranges for agricultural soils in the United States (Holmgren *et al.* 1993). Additionally, long-term organic amendments application at the rate used in this study did not increase extractable Cd, Cu, Pb, Ni, and Zn concentrations to a phytotoxic levels [e.g., >40, 50, and 400 $mg.kg^{-1}$ for Cu, Ni, and Zn (Chaney 1994)]. Plant toxicity was not observed nor were plant tissue levels elevated. These observations were consistent with other reports when tomatoes, squash, Chinese cabbage were grown in soils with organic amendments such as MSW and MSW/BS compost or high application rates of BS (Ozores-Hampton *et al.* 1994b and 1997; Wong 1996).

Tissue Analysis

No differences were observed in concentrations of heavy metals in pepper fruit in 1999 and 2000 with the exception of Zn (Table 5). Heavy metals in pepper fruit were lower than the maximum acceptable levels

TABLE 2.
Chemical analysis of organic amendments applied to the soil during 1993 to 2000 seasons

Property	MSW[z] Compost Broward County (1993)	Biosolids Tampa (1994)	YTW[y] Palm Beach (1995,1996,1997)	Cow Manure Compost (1996)	Biosolids Class B Miami (1998,1999,2000)
			(% Dry Weight)		
C	20.3	n/a[w]	26.7	6.5	36
N	1.2	3.0	1.9	0.34	5.7
P	0.3	2.3	1.0	0.44	2.7
K	0.4	0.13	0.5	0.28	0.14
Ca	3.1	1.65	4.1	0.90	6.0
			($mg.kg^{-1}$ Dry Weight)		
	3,000	1,900	2,721	2,300	8,345
Fe	20,000	39,000	10,401	1,136	13,150
Cd	2.9	0.7	3.0	n/a	7.2
Cu	281	662	161	16	627
Mn	5.8	3,180	111	68	40.0
Pb	231	53	60.2	n/a	98.0
Ni	34	92	8.2	n/a	153
Zn	655	700	266	97	1,395
			Additional Properties		
Moisture (%)	42.0	49.8	30.3	37.4	74.0
C:N ratio	16.9	n/a	14.5	19	6.4
pH	7.6	7.0	7.3	8.1	8.6
E.C. ($S m^{-1}$)	0.5	n/a	0.5	0.6	1.45

[z]MSW: Municipal solid waste, [y]YTW: Yard trimmings waste, [x]BS: Biosolids, [w]n/a: Not analyzed

TABLE 3.
Influence of yearly application of organic amendment on soil pollutant content over a four year-period

Treatments	Cd	Cu	Pb	Ni	Zn
	June 1996 (mg.kg^{-1})				
Organic amendment	0.008**z	7.8	0.478**	0.048**	22.3**
Nonamended control	0.0	6.7	0.040	0.010	4.7-
	June 1998 (mg.kg^{-1})				
Organic amendment	0.008**	10.5**	0.435**	0.056**	23.5**
Nonamended control	0.0	6.3	0.036	0.008	4.4
	June 1999 (mg.kg^{-1})				
Organic amendment	0.005*	15.0**	0.525**	0.085**	21.9**
Nonamended control	0.002	6.7	0.039	0.012	2.5
	June 2000 (mg.kg^{-1})				
Organic amendment	0.005	5.9	0.400*	0.075*	30.0**
Nonamended control	0.005	4.8	0.035	0.008	3.2

z**, *, Significant at P £ 0.01, P £ 0.05, respectively

TABLE 4.
Effects of yearly application of amendments in soil cumulative pollutant loading rate limits over a four-year period

	Cumulative Pollutant Loading Rate Year 2000 (Kg.ha^{-1})	Cumulative Pollutant Loading Rates Limits (Kg.ha^{-1}) Florida[z]	Cumulative Pollutant Loading Rates Limits [(Kg.ha^{-1}) Clean Water Act Section 503[x]]
Cd	0.011	5	39
Cu	13.2	124	1,500
Pb	0.89	499	300
Ni	1.17	124	420
Zn	67.3	249	2,800

[z]U. S. Environmental Protection Agency (USEPA) 1994 and 1995
[x]Department of Environmental Protection (DEP) 1989

TABLE 5.
Influence of yearly application of organic amendment on pepper fruit pollutant content over a two-year period.

Treatments	Cd	Cu	Pb	Ni	Zn
	Pepper Fruit 1999 (mg.kg^{-1})				
Organic amendment	0.005	0.475	0.015	0.020	0.675*z
Nonamended control	0.005	0.505	0.028	0.013	0.557
	Pepper Fruit 2000 (mg.kg^{-1})				
Organic amendment	0.005	0.478	0.023	0.025	0.602*
Nonamended control	0.005	0.543	0.035	0.023	0.482

z **, * Significant at $P \leq 0.01$, $P \leq 0.05$, respectively

for vegetables for Cd and Pb, 0.05 and 0.1 mg.kg^{-1}, respectively. Nickel, Cu and Zn were lower than the maximum levels found in pepper in the U.S, 0.21, 0.7, and 1.2 mg.kg^{-1} (USFDA 2000). Low pollutant loading rates and relatively high soil pH (6.7) were factors that limited accumulation of heavy metal pollutants in plant tissue (Ozores-Hampton *et al.* 1994a and b; Dixon *et al.* 1995). Similar results were obtained in research in Florida on tomatoes and squash grown on calcareous soil where biosolids, MSW, and biosolids-MSW compost that met the 503 standards, were applied. No trace metal accumulation was found in the edible plant parts (Ozores-Hampton *et al.* 1994a and b; and 1997).

Long-term additions of organic amendments such as biosolids and composted organic materials used in this experiment (at the rates tested) did not increase heavy metal loading rates nor increase their concentrations in pepper fruit. Thus, this practice can be considered safe in regard to pollution from heavy metals in Florida's sandy soils.

Acknowledgments

This research was supported in part by USDA - CSREES Regional IPM Grant No 39109813 and approved for publication as Florida Agricultural Experiment Station Journal Series No R- 09602.

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