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Heavy metal distribution in soil and plant in municipal solid waste compost amended plots

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ABSTRACT: A field study was carried out to evaluate long-term heavy metal accumulation in the top 20 cm of a Tunisian clayey loam soil amended for four consecutive years with municipal solid waste compost at three levels (0, 40 and 80 t/ha/y). Heavy metals uptake and translocation within wheat plants grown on these soils were also investigated. Compared to untreated soils, compost-amended soils showed significant increases in the content of all measured metals: cadmium, chromium, copper, nickel, lead and zinc in the last three years, especially for plots amended with municipal solid waste compost at 80 t/ha/y. Wheat plants grown on compost-amended soils showed a general increase in metal uptake and translocation, especially for chromium and nickel. This heavy metal uptake was about three folds greater in plots amended at 80 t/ha/y as compared to plots amended at 40 t/ha/y. At the end of the experimental period, the diluting effect resulting from enhanced growth rates of wheat plants due to successive compost applications resulted in lower concentrations in the plants (grain part) grown on treated plots. On the other hand, chromium and nickel were less mobile in the aerial part of wheat plants and were accumulated essentially in root tissues. Plant/soil transfer coefficients for compost-amended treatments were higher than threshold range reported in the literature, indicating that there was an important load/transfer of metal ions from soils to wheat plants.

Keywords: *Compost; Heavy metals; Metal uptake; Wheat plants*

INTRODUCTION

The concept of recycling waste nutrients and organic matter back to agricultural land is feasible and desirable. Land application represents a cost-effective outlet for the producers of compostable wastes and a potential cheap source of organic matter and fertilizer elements for landowners (Petruzzelli *et al.*, 1989; Chukwujindu, *et al.*, 2006; Nouri *et al.*, 2008).

Moreover, compost application to soil is used to maintain and improve soil structure (Paglia and Vittori-Antisari, 1993; Giusquiani *et al.*, 1995; Lillenberg, *et al.*, 2010) because its organic matter content can counteract the natural decline in intensively cultivated soils. It may even replace traditional farm manure whose availability in areas of intensive agriculture is often very poor. In addition to the potential beneficial nutrients, some waste materials may also contain non-essential elements, persistent organic compounds and microorganisms that may be harmful to plants (Kurihara, 1984; Mullin and Mitchell, 1994; Chukwuji *et al.*, 2005). For instance, the presence of toxic heavy metals in municipal solid waste composts (MSWC) raises serious concerns about the adverse environmental impact as a result of excessive application to agricultural lands (Nicholson *et al.*, 2003; Nouri *et al.*, 2006; Ayari *et al.*, 2008; Mahvi, 2008). Heavy metals originate mostly in non source-separated municipal solid wastes from a variety of sources: batteries, electronic appliances, newspapers, paint chips, foils, motor oils, and plastics that can all introduce metal contaminant into the compostable organic fraction (Hamdi *et al.*, 2003). High and excessive accumulation of heavy metals in soil and other media may eventually contaminate both human and animal food chain (Williams *et al.*, 1987; He *et al.*, 1992; Chukwuji *et al.*, 2005). For this reason, current European Community Regulations (1975 and

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1986) for land application of biowaste materials consider high content of heavy metals as a limiting factor for reuse purposes.

To correlate metal concentration in soil with plant uptake, a 4-year field study was undertaken: (a) to determine the accumulation of cadmium , chromium, copper, nickel, lead and zinc in the top 20 cm of an agricultural soil amended yearly with MSWC and (b) to determine the uptake of these metals and their distribution in wheat plants grown on this soil. To this end, a field-scale investigation was carried out from 1999 to 2002 at the agricultural experiment station of the Tunisia National Institute of Agronomy (INAT).

MATERIALS AND METHODS

Soil, MSWC and plant

The experiment station is located in northern Tunisia and belongs to the semi-arid superior bioclimatic stage with annual mean temperatures ranging between 15 and 29 °C, and average annual rainfall of 400-450 mm (Benzarti *et al*., 2007). The soil is loamy-clay classified as Xerofluvent with some physico-chemical properties illustrated in Table 1.

MSWC was obtained from the municipal composting plant of Henchir Lihoudia landfill, near Tunis the capital. The urban compost was produced under aerobic conditions by slow fermentation for about five months. MSWC maturity was monitored

Table 1: Soil physico-chemical characteristics in different soil depths

Parameter		0-20 cm	20-40 cm	40-60 cm	60-80 cm	80-100 cm	100-120 cm
Soil granulometric characteristics	coarse sand, %	1.85	0.82	1.24	1.25	1.58	1.59
	fine sand, %	3.42	3.47	4.63	3.8	7.11	7.72
	coarse silt, %	7.88	12.16	10.83	12.9	11.96	12.31
	fine silt, %	38.56	24.42	23.07	22.3	22.54	9.33
	clay, %	48.29	59.13	60.23	59.75	56.81	69.05
pH		8.74	8.75	9.08	9.16	9.12	8.84
OM, %		1.4	1.19	1.24	1.19	0.97	1.24
TOC, %		0.81	-	-	-	-	-
Total limestone, %		20	24	24	23.75	23	21.25
Active limestone,%		18.5	19	17	18.33	15.5	15.67
NH_4^+		6.85	8.61	8.05	6.28	7.28	6.67
NO_3^- NO_2^-		16.54	18.37	18.97	15.42	11.2	12.79
Total N, %		0.10	-	-	-	-	-
C/N		8.1	-	-	-	-	-
Assimilable P (ppm)		25.02	11.54	10.45	10.25	8.26	9.51
Total K (ppm)		5650	4550	4750	4350	4150	4500
Assimilable K (ppm)		440	286.67	240	206.67	220	213.33
Total Mg (ppm)		3380	3165	3190	3180	2910	2385
Assimilable Ca (ppm)		9650	9250	9300	8550	9793.33	9116.67
Total Na (ppm)		440	520	620	820	930	1040
Assimilable Na (ppm)		154.67	254.67	358.67	524	694.67	793.33
Total Zn (ppm)		94.8	98.25	96.85	93.85	86.95	94.8
Assimilable Zn (ppm)		1.54	1.06	1.78	1.45	1.77	2.3
Total Fe, %		0.21	0.12	0.18	0.18	0.16	016
Total Cu (ppm)		48.23	46.73	45.00	47.12	45.6	44.34
Total Zn (ppm)		92.12	90.34	92.10	91.4	90.43	90.00
Total Ni (ppm)		28.58	27.00	26.12	26.00	27.23	26.45
Total Cr(ppm)		32.41	31.51	30.24	29.21	31.30	29.00
Total Pb (ppm)		55.22	56.12	54.65	52.45	52.65	53.12
Total Cd (ppm)		1.10	0.99	0.94	0.88	0.91	0.89

by regular temperature measurements inside the waste pile and C/N ratio and decrease. Table 2 reports on some physico-chemical characteristics of the MSWC used in this study. Two levels of MSWC (40 and 80 t/ha/y) had been used yearly as autumnal amendment (1999-2000-2001-2002). Therefore, three soil treatments were named and distributed as follows:

T: Non-treated soil (control)

C1: MSWC added at 40 t/ha/y

C2: MSWC added at 80 t/ha/y

Table 2: Compost physico-chemical characteristics

Parameter	
pH	7.88
EC (µs/cm)	933
C, %	17.52
N, %	1.76
C/N	9.78
Cd (mg/kg)	5.17
Pb (mg/kg)	411.5
Cr (mg/kg)	78.87
Ni (mg/kg)	90.80
Cu (mg/kg)	337
Zn (mg/kg)	1174.5

All soil treatments were in four replicates. Plots were yearly cropped with irrigated durum wheat "cultivar Karim" (cv Karim) after MSWC amendments. After harvest (late June), surface soil (0-20 cm) and plant samples were collected from each plot for subsequent analyses.

Heavy metal analysis

Total heavy metals in soil and different parts of wheat plants were extracted by mineralizing 1 g of dry weight sample in concentrated HNO_3-HCl (1:3) for 12 h at ambient temperature, then for 8 h at 180 °C. Cd, Cr, Cu, Ni, Pb and Zn concentrations were determined by AAS and their content expressed as mg per g dw (Ayten, 2004).

Statistical analysis

Data were processed using STATISTICA 5.0 software and analyzed with ANOVA and *F*-test for mean separation.

RESULTS AND DISCUSSION

Table 3 reports on the variation of the total heavy metals content in top soil (0-20 cm) during 4 years of successive MSWC amendments.

At the end of the experimental period, Cd, Cr, Ni, Pb, Cu and Zn concentrations increased significantly in MSWC-amended soils as, compared to untreated control soils (Table 3). This increase was greater in soils amended with MSWC at the rate of 80 t/ha.

Table 3: Variation of total heavy metal concentration (mg/kg) in different soil treatments (0-20 cm) during 4 consecutive years

Year	Cd			Cr			Ni		
	T	C1	C2	T	C1	C2	T	C1	C2
1	1.05^a	1.06^a	1.06^a	34.21^a	36.64^a	40.06^a	29.90^a	30.54^a	32.80^a
2	0.88^a	1.15^a	1.17^a	34.05^a	40.53^a	41.70^a	25.74^a	28.63^a	31.29^a
3	1.05^a	1.31^a	1.56^b	33.62^a	41.85^a	49.95^b	29.58^a	36.82^b	43.94^b
4	1.00^a	1.96^b	2.51^b	35.02^a	52.96^b	64.15^b	29.26^a	40.89^b	57.13^c

Year	Pb			Cu			Zn		
	T	C1	C2	T	C1	C2	T	C1	C2
1	58.27^a	54.71^a	54.37^a	50.81^a	51.96^a	54.66^a	96.78^a	89.83^a	98.92^a
2	50.53^a	59.11^a	63.06^{ab}	48.67^a	54.95^a	57.10^a	88.78^a	96.63^b	100.3^a
3	57.02^a	70.97^b	84.71^{bc}	55.13^a	68.62^b	81.90^b	98.88^a	123.1^{bc}	146.9^b
4	66.64^a	105.2^c	126.4^c	51.01^a	73.58^b	89.37^b	92.31^a	151.4^c	176.3^c

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y

Column values with different superscripts for each element indicate a significant difference at $P < 0.05$

For instance, percent elevation for the six measured heavy metals after four amendments varied between 79 % for Cr to 130 % for Cd and Pb as compared to heavy metals content in the experimental soil. Repetitive addition of MSWC has been shown to inevitably increase heavy metals content in the surface layer of amended soils (Giusquiani *et al*., 1995; Crecchio *et al*., 2004; Madrid *et al*., 2006; Benzarti *et al.*, 2007; Nwachukwu *et al.*, 2010; Zaman, 2010). A parallel investigation on the same experimental but unplanted soils revealed that heavy metals and other potentially toxic elements content in soil greatly increased in the presence of MSWC after the second amendment (Yoshida *et al*., 2003; Benzarti *et al.*, 2007). Table 4 illustrates heavy metals concentration in the whole wheat plants grown on MSWC-amended plots and controls.The mean whole wheat plant dry weight increased continuously and proportionally to MSWC dose in the treated plots as result of beneficial soil enrichment with nutrients (Gallardo-Lara and Nogales, 1987; Hamdi *et al*., 2003, Castaldi *et al.*, 2005). Both Cd and Pb were not detected in plant tissues throughout the whole period of investigation (lower than 100 ppb for the analytical method used). As for MSWC-amended soils, measured heavy metals in plants were significantly greater than in control and were proportional to the level and number of MSWC amendments (Table 4). At the end of the experimental period, the content of heavy metals in plant tissues was the highest as compared to the previous sampling campaigns. This increase varied between 2 and 6-fold for Zn and Cr, respectively. The accumulation of Zn was the least as this element is known to be a mobile metal, easily leached by water infiltration in soil (Ayari *et al.*, 2008).

Total translocated Cr, Ni, Cu and Zn and their percent distribution in plant parts during the 4 years of MSWC amendment are presented in Tables 5-8. Absorbed heavy metals by wheat plants in the treated plots remained greater than those in untreated plots. For instance, Cr, Ni and Cu in plant tissues from plots treated with 80 t MSWC/ha/y were about 3-fold higher than in plants grown on soil amended at the rate of 40 t MSWC/ha/y. With a few exceptions, the general mean distribution of heavy metals in different plant parts was constant and was the same for plants grown with or without MSWC (Tables 5-8).

Cu and Zn, which are common micronutrients for the plants, were found in all plant parts including grains (Tables 7 and 8). The quantity absorbed by the plants from treated plots was higher, though their distribution was uniform in all plants from all treatments. Among the phytotoxic heavy metals studied in this research work (Ni, Cr and Pb), Ni was the most mobile (Kabata-Pendias and Pendias, 1992) although it was not detected in the grains (Hamdi *et al.*, 2003). On the other hand, toxic Cr was less mobile and accumulated mainly in root tissues (more than 70 %).

Although heavy metals concentration in MSWC-amended soils generally increased during the experimental period (Table 3), the total concentration of detected metals per plant tended to be stagnant from the second year of treatment because plant weight was enhanced by the fertilizing effect of the compost (Tables 5-8). The values obtained for heavy metals concentrations in the whole plant did not exceed the critical concentrations for plant growth and animal feed reported by Förstner (1995). Considering that the average weight distribution of stalks+leaves and grains were 41.8 % (± 1,5) and 48.2 % (± 1,3), respectively, the average concentrations of heavy metals in analyzed plant parts varied from 1.7 to 12.9 mg/kg for Cr, 2.1 to 14 mg/kg for Ni, 7.2 to 26.6 mg/kg for Cu and 66.8 to 118.7 mg/kg for Zn. These values are all below the lower limit of critical concentrations for animal feed. It can therefore be concluded that the long-term application of large amounts of urban waste compost to soils with similar physico-chemical characteristics does not necessarily cause medium-term problems to plant, animal and human health.

The plant/soil transfer coefficients (Kloke *et al.*, 1984) are shown in Table 9. Transfer coefficient (TC) is obtained from the ratio of each detected metal concentration in fresh plant tissues to that in soil. The TC quantifies the relative differences in bioavailability of soil metal to plant and is a function of both soil and plant properties. Control soils showed TC below the range of threshold values reported in literature (Förstner, 1995), testifying to the absence of heavy metals accumulation risks within plant tissues. In presence of MSWC, TC of Cr, Ni and Cu were higher than standard values counting from the second year of the biowaste amendment, indicating that there was an important load/transfer of metal ions from amended soils to wheat plants. TC values were always positively

Table 4. Variation of whole plant dry weight and plant content in heavy metal during 4 consecutive years

Year	Dry wt. (mg)			Heavy metals (mg/kg) Cr			Ni			Cu			Zn		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	90.5[a]	110.2[a]	130.3[a]	0.39[a]	1.00[a]	2.87[a]	0.23[a]	1.16[a]	3.04[a]	1.83[a]	4.18[a]	7.12[a]	19.77[a]	36.78[a]	53.34[a]
2	120.3[a]	150.5[ab]	200.6[b]	0.66[a]	4.61[b]	10.87[b]	0.66[a]	4.41[b]	10.34[b]	1.77[a]	11.51[b]	25.26[b]	18.43[a]	51.38[b]	71.65[b]
3	100.2[a]	198.4[b]	250.4[b]	0.63[a]	5.39[b]	12.10[b]	0.68[a]	5.63[b]	12.46[b]	1.68[a]	15.32[b]	33.34[b]	18.41[a]	62.75[b]	77.05[b]
4	96.5[a]	211.0[b]	253.2[b]	0.60[a]	5.95[b]	13.35[b]	0.67[a]	6.20[b]	13.70[b]	1.75[a]	16.89[b]	36.72[b]	18.36[a]	66.78[b]	82.05[b]

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y
Column values with different superscripts for each element indicate a significant difference at $P < 0.05$

Table 5: Chromium content and distribution in different plant parts during 4 consecutive years

Year	Total Cr (mg/plant)			Distribution in plant parts (%) roots			(stalks + leaves)			grains		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	0.04	0.11	0.37	86.51	65.78	80.56	13.49	34.22	19.44	ND	ND	ND
2	0.08	0.69	2.18	64.11	66.51	66.51	35.89	33.49	33.49	ND	ND	ND
3	0.06	1.07	3.03	63.62	65.11	67.91	36.38	34.89	32.09	ND	ND	ND
4	0.06	1.26	3.38	61.80	64.82	67.79	38.20	35.18	32.21	ND	ND	ND
Average	0.06	0.78	2.24	69.01	65.55	70.69	30.99	34.44	29.30	ND	ND	ND

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y
ND: Not detected

Table 6: Nickel content and distribution in different plant parts during 4 consecutive years

Year	Total Ni (mg/plant)			Distribution in plant parts (%) roots			(stalks + leaves)			grains		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	0.02	0.13	0.40	72.00	70.33	77.06	28.00	29.67	22.94	ND	ND	ND
2	0.08	0.66	2.07	60.05	65.11	65.44	39.95	34.89	34.56	ND	ND	ND
3	0.07	1.12	3.12	54.57	61.69	65.92	45.43	38.31	34.08	ND	ND	ND
4	0.06	1.31	3.47	51.75	61.61	66.01	48.25	38.39	33.99	ND	ND	ND
Average	0.06	0.80	2.26	59.59	64.68	68.60	40.41	35.32	31.40	ND	ND	ND

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y
ND: Not detected

Table 7: Copper content and distribution in different plant parts during 4 consecutive years

Year	Total Cu (mg/plant)			Distribution in plant parts (%) roots			(stalks + leaves)			grains		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	0.17	0.46	0.93	26.98	42.85	50.78	43.50	39.04	26.44	29.53	18.11	22.78
2	0.21	1.73	5.07	33.92	41.78	44.92	31.47	35.61	37.89	34.61	22.60	17.20
3	0.17	3.04	8.35	38.42	47.70	51.72	32.57	31.34	31.55	29.01	20.96	16.73
4	0.17	3.56	9.30	38.23	47.53	51.71	32.74	31.59	31.69	29.03	20.88	16.60
Average	0.18	2.20	5.91	34.38	44.96	49.78	35.07	34.39	31.89	30.54	20.63	18.32

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y

Table 8: Zinc content and distribution in different plant parts during 4 consecutive years

Year	Total Zn (mg/plant)			Distribution in plant parts (%) roots			(stalks + leaves)			grains		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	1.79	4.05	6.95	33.47	39.52	45.63	21.39	23.08	27.95	45.14	37.40	26.42
2	2.22	7.73	14.37	27.41	36.52	38.66	21.74	28.85	30.63	50.85	34.64	30.71
3	1.84	12.45	19.29	29.13	41.35	37.08	22.66	26.51	32.63	48.22	32.13	30.29
4	1.77	14.09	20.78	29.82	40.75	36.61	23.50	26.99	33.13	46.68	32.26	30.26
Average	1.91	9.58	15.35	29.96	39.54	39.50	22.32	26.36	31.09	47.72	34.11	29.42

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y

Table 9: Plant/soil transfer coefficients for absorbed metals during 4 consecutive years

Year	Cr			Ni			Cu			Zn		
	T	C1	C2	T	C1	C2	T	C1	C2	T	C1	C2
1	0.01	0.03	0.07	0.01	0.04	0.09	0.04	0.08	0.13	0.20	0.41	0.54
2	0.02	0.11	0.26	0.03	0.15	0.33	0.04	0.21	0.44	0.21	0.53	0.71
3	0.02	0.13	0.24	0.02	0.15	0.28	0.03	0.22	0.41	0.19	0.51	0.52
4	0.02	0.11	0.21	0.02	0.15	0.24	0.03	0.23	0.41	0.20	0.44	0.47
Range	0.01-0.1			0.01-0.1			0.01-0.1			1-10		

T: Control soil (unamended plots); C1: 40 t MSWC/ha/y; C2: 80 t MSWC/ha/y

proportional to the level of MSWC in soil for 4 consecutive years with the following order: C2 > C1 > T (Table 9). On the other hand, plant/soil TCs in MSWC-amended plots significantly increased after the second amendment before stabilization in the case of Ni and Cu or even a decrease in the case of Zn (Table 9).

CONCLUSION

Trace metal composition of MSWC-amended soil is an important factor to estimate total trace metal uptake by plants and consequently the level of risk of these elements for plants and food chain. In this study, heavy metals accumulation in the top 20 cm of a Tunisian clayey loam soil amended with MSWC for four consecutive years at the rate of 40 or 80 t/ha/y was evaluated. By time, the content of measured heavy metals significantly increased in the MSWC amended plots. Wheat plants grown on MSWC-amended soils showed also a general increase in metal uptake and translocation. However, only Zn and Cu were detected in the edible parts for humans (grains). On the other hand, Cr and Ni were less mobile and were accumulated essentially in root tissues. Transfer coefficients increased with the level of MSWC in soil for 4 years reflecting the important load/transfer of heavy metals in the soil/plant system.

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