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## Impact of Municipal and Industrial Waste on the Distribution and Accumulation of Some Heavy Metals in Sandy Soils of Al-Qassim Region at Central of Saudi Arabia

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**Abstract:** An experiment was conducted to study the distribution and accumulation of Pb, Ni, Co, Cu, Zn, Mn and Fe throughout sandy soils treated with industrial wastes. Surface and subsurface soil samples were collected from three sites namely; Aldahy, Almota and Alsenayh, which representing the major agricultural soils of Qassim, central region of Saudi Arabia. These sites have been used for dumping domestic and industrial wastes for more than 20 years to evaporate and infiltrate solid and liquid industrial waste. The obtained results indicated that, the application of industrial wastes resulted in a marked increase in the total amount of studied metals. Maximum concentrations of metals were found in the surface 30 cm of the soil where Fe > Mn > Zn > Ni > Pb > Cu > Co in most cases. Most movement and amount of metals recovered from soil were predominantly limited to the upper 30 cm depth. The DTPA fraction of metals was relatively small comparing either to the total soil content and/or the 0.5 N HNO<sub>3</sub> extractable fractions. For example, the average DTPA extractable Pb was highest (3.37 µg g<sup>-1</sup>) in Almota soils, 2.41 µg g<sup>-1</sup> Aldahy soil and 1.08 μg g<sup>-1</sup> in Alsenayh soil. The respective Pb values extracted by 0.5 N HNO<sub>3</sub> for the three sites were 6.75, 5.37 and 4.49  $\mu$ g g<sup>-1</sup>. Total Pb values were 23.23, 21.77 and 17.63 ug g<sup>-1</sup>, respectively. All the three forms were highest in Almota area and lowest in Alsenayh area. The average Ni content of the three soils was more or less in the same range as Pb. However, total Ni values were 2 to 3 times higher in Alsenayh soils. Though, these values were still low as compared to the contaminated reported levels but it indicated that these have been contaminated with the dumping of industrial waste. The values of Co for DTPA, HNO<sub>3</sub> and total extracts were relatively low when compared to Pb and Ni. Average DTPA content was 0.65 µg g<sup>-1</sup> for Alsenayh while it was 0.64 µg g<sup>-1</sup> for Aldahy and 2.02 µg g<sup>-1</sup> for Almota soils. The data shows a high accumulation of these metals in contaminated area. Accumulation rate was highest for Ni, Cu, Co, Pb followed by Fe, Zn and Mn.

**Key words:** Heavy metals, movement, accumulation mobility, municipal and industrial wastes

## INTRODUCTION

Increasing waste production by human activities has increased the problem of how to get rid of it without causing undesirable impact on the environment and human health. Traditional approaches, incineration and land filling, for remediation of toxic heavy metal contaminated soils are typically expensive, labor intensive and environmentally inefficient. In principal land disposal is more popular than other means including landfills, incineration and dumping at sea, because it is relatively inexpensive. Unfortunately, land disposal of these wastes is a potential risk for human safety due to inorganic pollutants such as heavy metals borne in these wastes. Concern about metal contamination of soils relates directly to the extent to which natural background levels are exceeded and to the mobility of these metals in terms of bioavailability to plants and the movement to ground water.

It has been generally assumed that since heavy metals are strongly bound by the soil, leaching of metals downward into ground water is not an environmental issue. However, in recent years, this assumption is challenged and metal leaching from sludge treated soil has been observed in several studies. Heavy metals are associated with various soil components in different ways and these associations indicate their mobility in soils as well as their bioavailability (Al-Oud, 2000; Al-Wabel and El-Maghraby, 2006). The degree of metal association with different chemicals forms depends on soil properties such as pH, organic matter, redox condition and soil texture (Zhang *et al.*, 1997).

Land application of bio-solids and industrial wastes can significantly increase heavy metals concentrations in agricultural soils. Fields studies have generally shown no difference in heavy metals concentration below 30 cm between treated and untreated soils with bio-solids (Chang et al., 1982, 1983; Dowdy and Volk, 1983; Williams et al., 1985; Al-Oud, 2003; Zheljazkov and Warman, 2004). However, leaching of some heavy metals to 60 cm depth can occur when large amounts of liquid sewage sludge are applied (Robertson et al., 1982). Fuller (1997) reported Cd to be fairly mobile in soils of pH 4.6 to 6.6 and moderately mobile in soils with pH range 6.7-7.8. Sanchez-Martin and Sanchez-Camazano (1993) in their study concluded that Cd was slightly mobile in 27%, moderately mobile in 14%, mobile in 41% and highly mobile in 18%. They further reported that, the pH, exchangeable bases and clay content of soil have a significant effect on Cd mobility. Scokart et al. (1983) observed higher accumulation of Cd and Zn in loamy soils than for sandy soils even though they have the same CEC. According to Elliot and Denneny (1982), the addition of organic rich waste such as municipal sludge and industrial wastes can provide enough organic matter to the soils to bind toxic heavy metals like Cd, Zn and Pb. Also Kabala and Singh (2001) and Zheljazkov and Warman (2004) reported that organic matter had high ability to absorb or complex many metals. In addition, adsorptions on hydrous oxides of iron and cation exchange reactions are important modes of metal immobility (Pilar et al., 2006). In most cases, the metals are present in an immobile form at the time they are added to the soil. Heavy metals are associated with various soil components in different ways and these associations indicate their mobility in soils as well as their bioavailability. Tania et al. (2003) and Zheljazkov and Warman (2004) pointed out that, hydrosoluble and exchangeable fractions are considered readily mobile and bioavailable.

A large number of articles on heavy metals concentration in agricultural soils can be found in the literature, however, elemental background or reference values in uncultivated soil are limited (Angelone and Bini, 1992; Chen *et al.*, 1991; Lena *et al.*, 1997). Knowledge on this topic is scare with a shortage in the information in the Kingdom of Saudi Arabia. Therefore, the present research was carried out with the objective to evaluate contamination degree of heavy metals and their mobility in some major agricultural soils of Al-Qassim region due to human activities and industrial waste disposal.

## MATERIALS AND METHODS

In 15 May, 2007, surface and subsurface soil samples, were collected from three sites namely, Aldahy, Almota and Alsenayh, which representing the major agricultural soils of Qassim, central region of Saudi Arabia. These soils are classified as (Torriorthent) according to the soil taxonomy of the Soil Survey Staff (1999). These sites have been used for dumping domestic and industrial wastes. Site 1 (Almota) is industrial waste, home garbage and auto wrecks. This site used by Buriedah city as a landfill from early 1950s until late 1970s. The land surrounding this site is a semi-rural, residential and farming field. Site 2 (Aldahy) used to dispose excess rain from the Buriedah city streets and overflow of municipal water form late 1960s to late 1970s. The land surrounding this site is residential community. Site 3 (Alsenayh), is being used since late 1970 until now to evaporate and infiltrate solid and liquid industrial waste. It is mainly surrounded by factories and farming fields. Sixteen soil samples representing four profiles were collected from Alsenayh site, to study the mobility of heavy metals in this area which is being dumped with industrial city waste.

Table 1: General characteristics of the studied soils

Soil properties	Alsenayh soil	Aldahy soil	Almota soil
Sand (%)	83.80	78.30	84.30
Silt (%)	6.60	11.30	7.40
Clay (%)	9.60	10.40	8.30
pH 1:1	8.12	8.32	8.41
OM (%)	1.22	0.38	0.55
CaCO <sub>3</sub> (%)	3.40	2.25	4.25
EC (dS m <sup>-1</sup> )	6.05	3.79	4.20

Soils were air dried, passed through a 2 mm sieve and stored for analysis. Particle size distribution was determined by the hydrometer method (Gee and Bauder, 1996). Content of CaCO<sub>3</sub> was determined by calcimeter method (Loeppert and Suarez, 1996). Soil organic matter of the soil samples were determined by digested them using concentrated H<sub>2</sub>SO<sub>4</sub> (Nelson and Sommers, 1996). Soil pH and EC were measured in distilled water extracts after equilibration for 24 h. The ratio of soil and distilled water was 1:1 (Thomas, 1996; Rhoades, 1996). Table 1 shows some physical and chemical properties of the studied soils.

Heavy metals (Pb, Ni, Co, Cu, Zn, Mn and Fe) were extracted by DTPA as described by Lindsay and Norvell (1978). Acid extractable metals were determined using 0.5 N HNO<sub>3</sub> as described by Sloan *et al.* (1997). Moreover the total content of heavy metals soils were digested by using a mixture of concentrated HClO<sub>4</sub> and HNO<sub>3</sub> at a 2:1 ratio and then dissolving the extracts in 0.5 N HCl. The metals in all the three extracts were determined using ICP spectroscopy, model GBC-XI, Integra. All the analysis was doubled read for each sample and the data presented in this paper are the average of three replicates of each sample.

#### RESULTS AND DISCUSSION

## **Accumulation of Heavy Metals**

Average DTPA extractable Pb content was highest  $(3.37~\mu g~g^{-1})$  in Almota site, 2.41 Aldahy site and 1.08 in Alsenayh site. The respective Pb values extracted by HNO $_3$  for the three sites were 6.75, 5.37 and 4.49  $\mu g~g^{-1}$ . Total Pb values were 23.23, 21.77 and 17.63  $\mu g~g^{-1}$ , respectively (Table 2 A, B) All the three forms were highest in Almota site and lowest in Alsenayh site. When these values were compared with the values (10-100  $\mu g~g^{-1}$ ) reported by Alloway (1990) fall in critical range.

The average Ni content of the three soils was more or less in the same range as Pb. However, total Ni values were 2 to 3 times higher in three Alsenayh soils. Though, these values were still low as compared to the contaminated reported levels but it indicated that these have been contaminated with the dumping of industrial waste. The values of Co for DTPA, HNO<sub>3</sub> and total extracts were relatively low when compared to Pb and Ni average DTPA content was  $0.65~\mu g~g^{-1}$  for Alsenayh  $0.64~\mu g~g^{-1}$  for Aldahhy and higher  $2.02~\mu g~g^{-1}$  for Almota soils. The respective, average content of Cu extracted by HNO<sub>3</sub> in the three sites were 1.42, 1.77 and  $3.20~\mu g~g^{-1}$ . Total average values for Cu were 6.8,  $12.25~and~19.16~\mu g~g^{-1}$ , respectively. All the three forms were highest in Almota area and lowest in Alsenayh area. DTPA extractable Zn values were some what higher in all the three soils (mean values were 7.5,  $5.47~and~7.37~\mu g~g^{-1}$ ) reaching to as high as  $19.82~\mu g~g^{-1}$  in soil no. 1 of Alsenayh.

The respective Zn values extracted by  $\rm HNO_3$  for the three sites were 16.38, 30.50 and  $28.2~\mu g~g^{-1}$ . Total Zn values were 35.53, 92.98 and  $122.36~\mu g~g^{-1}$ , respectively, respectively. It seems from the results that Zn is being added to soil from outer source. The reported content of Zn extracted by AB-DTPA in previous studies ranged 0.14- $7.46~\mu g~g^{-1}$  with mean value  $1.44~\mu g~g^{-1}$  (Ayed and Choudhary, 1989) while Bashour *et al.* (1983) reported the similar results (0.2-3.2  $\mu g~g^{-1}$ ) when Zn was extracted with DTPA. High values found in our soils gave a clear indication that soils are contaminated as the sites were used for dumping of wastes. Content of DTPA and HNO<sub>3</sub> extractable

Table 2A: Content of different forms of heavy metals (µg g<sup>-1</sup>) extracted from the experimental sites

		Pb			Ni			Со		
Location	Depth (cm)	A	В	С	Α	В	С	Α	В	C
Alsenayh site	0-30	0.54	12.10	18.33	0.96	2.65	50.00	0.17	1.65	2.74
•	0-30	0.53	2.90	19.90	1.10	3.80	48.75	0.21	1.25	2.19
	0-30	1.60	2.95	15.74	1.83	1.90	36.50	0.33	1.00	2.08
	0-30	1.02	1.65	11.50	1.62	2.70	38.75	0.07	1.05	2.65
	0-30	1.22	1.85	15.75	1.38	1.35	59.25	0.58	1.70	3.50
	0-30	2.59	5.50	14.50	0.98	1.55	64.50	0.12	1.50	4.59
	Mean	1.25	4.49	15.95	1.31	2.33	49.63	0.25	1.36	2.96
Aldahy site	0-30	2.10	1.95	27.00	1.57	1.20	6.50	0.30	1.60	2.65
<u> </u>	0-30	2.05	1.80	12.82	1.12	1.55	4.93	0.11	1.35	3.08
	0-30	2.75	12.35	25.50	1.05	1.75	5.75	0.08	1.70	3.90
	Mean	2.30	5.37	21.77	1.25	1.50	5.73	0.16	1.55	3.21
Almota site	0-30	3.71	8.75	21.25	1.24	1.60	17.00	0.11	1.70	4.53
	0-30	4.95	6.25	38.50	1.50	1.45	17.50	0.15	1.75	5.84
	0-30	3.47	5.25	9.95	1.86	1.40	17.00	0.05	1.55	3.03
	Mean	4.04	6.75	23.23	1.53	1.48	17.17	0.10	1.67	4.47

A: Available forms extracted with DTPA, B: Mobile forms extracted with  $0.5~N~HNO_3$ , C: Total heavy metals extracted with a mixture of concentrated HClO $_4$  and HNO $_3$  at a 2:1 ratio and then dissolving the extracts in 0.5~N~HCl

Table 2B: Content of different forms of heavy metals (ug g<sup>-1</sup>) extracted from the experimental sites

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		Cu			Zn			Mn			Fe		
	Depth												
Location	(cm)	A	В	С	Α	В	С	A	В	C	A	В	C
Alsenayh site	0-30	0.88	2.00	4.50	19.82	40.80	44.00	11.73	19.60	77.25	34.89	831.00	5280
	0-30	0.91	0.35	4.25	11.20	24.80	32.45	2.98	11.60	68.50	49.72	705.00	5990
	0-30	0.03	0.06	3.50	1.96	3.50	13.25	2.69	11.35	75.80	26.62	462.50	5230
	0-30	1.09	0.03	6.00	9.50	20.04	33.00	8.91	24.60	145.00	34.94	431.00	6710
	0-30	0.75	0.08	16.00	1.81	6.60	53.00	8.13	9.20	280.00	1.69	13.55	6300
	0-30	0.22	0.03	6.75	0.92	2.52	22.10	1.80	18.20	239.80	1.64	14.60	6800
	Mean	0.65	0.43	6.83	7.54	16.38	32.97	6.04	15.76	147.73	24.92	409.61	6051
Aldahy site	0-30	0.43	0.03	13.88	1.39	5.10	41.50	7.44	9.95	255.00	7.28	38.50	19600
	0-30	0.30	1.10	7.58	2.55	39.60	59.73	2.41	22.75	203.40	1.61	43.60	12600
	0-30	1.19	2.20	15.30	12.49	46.80	177.70	7.01	45.02	258.20	5.01	43.70	12400
	Mean	0.64	1.11	12.25	5.48	30.50	92.98	5.62	25.91	238.87	4.63	41.93	14866
Almota site	0-30	1.79	1.10	20.88	6.83	41.16	97.25	6.17	28.35	168.60	3.82	35.40	15400
	0-30	3.66	0.30	29.50	13.30	41.20	248.70	7.90	22.65	198.30	4.51	27.50	14600
	0-30	0.62	0.20	7.10	1.97	2.44	21.13	5.70	21.85	102.90	4.24	19.60	9900
	Mean	2.02	0.53	19.16	7.37	28.27	122.36	6.59	24.28	156.60	4.19	27.50	13300

A: Available forms extracted with DTPA, B: Mobile forms extracted with 0.5 N HNO $_3$ , C: Total heavy metals extracted with a mixture of concentrated HClO $_4$  and HNO $_3$  at a 2:1 ratio and then dissolving the extracts in 0.5 N HCl

Mn were same as Zn. However, total average contents were 147.72, 238.87 and 156.6  $\mu$ g g<sup>-1</sup>, respectively for Alsenaeyh, Aldahy and Almota sits. DTPA extractable Fe was very high in Alsnaeyah with an average of 24.92  $\mu$ g g<sup>-1</sup> reaching up to 49.72  $\mu$ g g<sup>-1</sup>. HNO<sub>3</sub> content for the soil soils was 409.6  $\mu$ g g<sup>-1</sup>.

The data shows a high accumulation of such metals in contaminated area. Accumulation rate was highest for Ni, Cu, Co, Pb followed by Fe, Zn and Mn. Though all these metals contents were well in normal range (Kabata-Pendias and Pendias, 1992; Alloway, 1990) in natural soils but their concentrations has increased to critical levels according to criteria laid by Alloway (1990). Ni has touched to 218 mg kg $^{-1}$  while toxic level was put > 100 mg kg $^{-1}$  by Kabata-Pendias and Pendias (1992).

## **Movement of Heavy Metals**

In all the four profile DTPA extractable as well as 0.5 N HNO<sub>3</sub> digested soils (Table 3A, B) for all the metals studied were highest in the surface soils (0-30 cm) and their concentrations decreased

Table 3A: Distribution of different forms of heavy metals (μg g<sup>-1</sup>) extracted from Alsenayh soils

		Pb	-		Ni		-	Со		
Profile No.	Depth (cm)	A	В	С	A	В	С	Α	В	С
1	0-30	0.1	2.45	19.6	2.46	3.75	120	0.11	2.15	2.56
	30-60	0.06	1.60	8.9	1.65	1.70	84	0.03	0.95	2.29
2	0-30	0.31	1.85	23.0	1.78	3.52	218	0.14	1.35	2.56
	30-70	0.29	2.20	21.3	0.03	2.35	158	0.10	1.30	2.53
	70-100	0.11	10.0	17.4	0.98	1.75	130	0.06	0.85	1.84
3	0-20	2.5	5.25	49.8	2.68	8.15	185	0.79	1.80	1.89
	20-40	0.62	5.20	28.3	1.34	1.95	169	0.67	1.10	1.69
	40-60	0.71	2.25	27.0	1.91	1.95	150	0.62	1.15	1.65
	60-80	0.14	1.85	14.0	1.09	1.85	125	0.62	1.10	1.63
	80-100	0.02	1.50	8.3	0.93	1.60	123	0.55	1.00	1.60
4	0-20	0.33	0.25	21.8	2.13	3.55	74	0.60	1.55	11.10
	20-40	1.48	2.10	15.3	1.15	2.30	74	0.52	1.50	5.00
	40-60	0.63	0.95	9.7	1.00	2.60	67	0.50	1.30	3.60
	60-80	0.44	3.30	7.3	0.56	2.55	61	0.45	1.15	2.80
	80-100	0.41	2.89	6.0	0.37	1.60	43	0.22	1.10	1.30
	100-120	0.26	0.75	3.5	0.29	2.65	32	0.12	0.42	0.85

A: Available forms extracted with DTPA, B: Mobile forms extracted with  $0.5 \, N \, HNO_3$ , C: Total heavy metals extracted with a mixture of concentrated  $HClO_4$  and  $HNO_3$  at a  $2.1 \, ratio$  and then dissolving the extracts in  $0.5 \, N \, HCl$ 

Table 3B; Distribution of different forms of heavy metals (ug g<sup>-1</sup>) extracted from Alsenayh soils

		Cu			Zn			Mn			Fe		
	Depth												
Profile No	. (cm)	A	В	С	A	В	C	Α	В	С	A	В	С
1	0-30	0.80	2.23	4.5	0.72	4.20	10.3	4.30	65.2	110.0	18.7	739.0	8900
	30-60	0.02	0.62	3.75	0.52	2.45	8.1	3.53	31.5	100.0	16.5	291.5	7100
2	0-30	0.43	1.20	4.58	0.62	1.91	15.4	3.10	48.2	137.5	16.8	1010.5	7700
	30-70	0.03	0.37	3.25	0.85	2.80	13.1	1.80	29.6	125.0	2.57	240.5	3900
	70-100	0.02	0.19	3.05	0.53	2.30	12.7	0.98	26.1	80.0	1.13	172.5	3200
3	0-20	2.19	9.75	14.00	49.10	114.40	49.4	24.00	46.0	110.0	44.1	950.0	7600
	20-40	0.07	1.03	6.38	1.52	4.10	15.4	17.40	31.1	78.5	26.1	715.0	6600
	40-60	002	0.92	5.75	1.40	3.05	14.3	16.74	29.6	73.0	26.0	697.0	6300
	60-80	0.02	0.88	4.00	1.39	2.30	12.4	12.90	23.1	74.03	19.1	679.0	5500
	80-100	0.02	0.39	2.80	1.13	2.05	12.1	5.20	11.1	72.5	16.7	325.0	4500
4	0-20	0.75	1.35	4.50	1.58	3.43	21.8	4.62	69.0	196.5	25.9	914.5	10100
	20-40	0.03	0.60	4.10	1.22	6.00	19.8	4.40	37.6	158.5	17.1	178.5	10400
	40-60	0.02	0.25	3.00	0.98	3.05	18.4	2.72	31.2	149.5	14.6	175.5	9900
	60-80	0.02	0.18	2.25	0.48	1.90	15.7	2.04	21.1	143.4	11.0	161.0	7900
	80-100	0.01	0.15	1.73	0.44	1.78	15.0	1.69	17.8	79.5	6.09	117.5	5400
	100-120	0.01	0.05	0.48	0.29	1.70	11.1	1.67	14.9	30.0	5.12	123.0	4800

A: Available forms extracted with DTPA, B: Mobile forms extracted with 0.5 N HNO $_3$ , C: Total heavy metals extracted with a mixture of concentrated HClO $_4$  and HNO $_3$  at a 2:1 ratio and then dissolving the extracts in 0.5 N HCl

with depth. Profile No. 3 and 4 have much higher concentrations of Zn, Mn and Fe both in surface and subsurface soils. Such findings were stood in agreement with those of and (Zheljazkov and Warman, 2004; Pilar *et al.*, 2006; Sukkariyah *et al.*, 2005a, b, 2007). The other hand, the concentrations of Fe seem to be high however, it is still in the common ranges in the soils found by Lindsay (1979). Higher content of these metals may create a risk for contamination of agricultural crops grown in the surroundings. These findings were in agreement with Kashem and Singh (2002) who reported Cd highest in the surface layer and decreased from A to C horizon. Many other researchers have found no difference in heavy metals concentrations below 30 cm between treated and untreated soils with bio-solids (Chang *et al.*, 1982, 1983; Dowdy and Volk, 1983; Williams *et al.*, 1985; Zhang *et al.*, 1997; Pilar *et al.*, 2006; Sukkariyah *et al.*, 2007). There was little evidence of downward movement of Ni, Co and Cu. These soils are calcareous in nature and probably most of the metals may precipitate with calcium carbonate and also due to a high pH value about 8 most of heavy metals may precipitate in

their hydroxide form. Therefore restricts their mobility to the lower depth. However, if heavy metals contamination exceeds the binding capacity of the soil, there is a danger that heavy metal compounds can percolate into the groundwater. For Pb only mobile form extracted by 0.5 N HNO<sub>3</sub> showed different pattern of distribution. It did not follow the decreasing pattern. Profile 2 and in particular profile 4 there was 2-3 fold higher Pb concentration at 60-80 cm and 80-100 cm depth. This shows that Pd is more susceptible to mobility than other elements. For total Pb it decreased with depth similar to DTPA extractable Pb. Also, DTPA, 0.5 N HNO<sub>3</sub> and Total Ni content though high but their concentrations decreased with depth. Total Ni content as compared to the normal soils however; it was accumulated only on the surface 30 cm depth of Alsenayh soil.

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