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ENRICHMENT FACTOR OF HEAVY METALS IN DIFFERENT SOIL GRAIN SIZE FRACTIONS AS AN INDICATOR FOR SOIL POLLUTION

F. H. Rabie, M. F. Abdel-Sabour*, A. T. Mostafa and S. A. Hassan**

Soil Department, Faculty of Agricultural, Ain Shams University, *Soil and Water Pollution Unit, Nuclear Research Center, **Soils, Water and Environment Research Institute, ARC, Soils, Egypt.

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ABSTRACT :

An industrial area north of greater Cairo was selected to investigate the impact of intensive industrial activities on soil characteristics and heavy metals content. The studied area was divided into six sectors according to its source of irrigation water and/or probability of pollution. Sixteen soil profiles were dug and soil samples were taken, air dried, fractionated to different grain size fractions, then total heavy metals (Zn, Cu, Co, and Cd) were determined using ICP technique. The enrichment factor for each element, for each soil fraction/soil layer was estimated and discussed. Results revealed that the increase of EF-values with soil depth, in some cases, may suggest that some heavy metals in the contaminated soils are mobile and leacheable through the soil profile due to the presence of organic complexes usually found in wastewater effluent. These dissolved organic compounds act as chelating agents and protect metals from the ordinary precipitation and fixation process in arid soils. The highest EF ratios in the clay fraction were mainly with Cd which may shows the industrial impact on the soil. Concerning silt fraction, a varied accumulation of Zn and Cu was observed with soil depth and different soil profiles

INTRODUCTION :

In addition to the natural constituents, heavy metals may enter the soil via atmospheric deposits, discharges from sewage treatment plants, application of sewage sludge as fertilizer or landfill material, polluted irrigation water and beneficial agricultural additives. The soils become polluted if metals content were above an accepted normal range and may affect the growing crops.

Knowledge of soil total content of heavy metals provides only limited information as this can seldom be correlated with availability to plants and does not show how the metal is bound in the soil. In both pedological and environmental studies, it would be helpful to know the forms in which heavy metals occur, and also the propotions of these forms in the soil. The distribution of these metals in particle size fraction is one of these forms [1]. A number of authors have fractionated soil in order to determine the relative contributions of clay, silt and sand to heavy metal content of a certain soil. Nair and Cottenie [2] found that Zinc, copper and lead are bound up in the clay and /or silt fractions.

The total metal content of the soil and metal content of each fraction were used to calculate the enrichment factor for each fraction at different soil layers [3]. This factor could be a useful diagnostic tool to asses the environmental impact of industrial wastewater irrigation on agri-cultural land [4]. It is indicated that the continuous irrigation with wastewater will lead to the enrichment of these soils by heavy metals. Banin et al [5] reported that continuous irrigation with sewage effluent resulted in increasing Cd, Cu, Ni and Pb metals in the surface and subsurface layers of soil profile. Hemkes et al, [6] reported that the mobility of the metal ion would determine the rate of penetration to the subsoil layer.

The aim of this study is to asses soil enrichment by heavy metals due to waste- water irrigation in an industrial area north greater Cairo, Egypt.

EXPERIMENTAL :

Materials and Methods :

The studied area is an industrial area north of greater Cairo region. It encompasses a strange mixed human activities (e.g. housing, agricultural and different types of industrial activities). The area suffers from several environmental problems such as industrial wastewater and sewage effluent disposal. Some of the agricultural fields are irrigated by wastewater directly without any treatment. The analysis of these waters showed that El-Shaboura canal and Shebin El-Qanater collector drain exhibited the highest levels of Cd, Co, Pb and Ni while, Mostorod collector drain has the highest levels of Zn and Cu [7] . The area was divided into six sectors according to the source of irrigation water and the probability of pollution with organic and inorganic wastes (Fig. 1). Sixteen soil profiles were dug and soil samples were taken from different layers. Another soil samples were

taken from a virgin soil at the vaccinity to represent uncultivated control soil (Sector A) which is represented by soil profile No. 1 .

Sector B: the source of irrigation water for soils of this sector is regular fresh water from El-Boulaqeyah canal which is located far from the pollution sources in the studied area. These soils are considered non-polluted and are taken as control. Profile No. 2 represent this sector.

Sector C: The soils of this sector are subjected to irrigation with heavily polluted wastewater from Shebin El-Qanater collector drain (receives wastewater from plastic, staining and fabric factories), profile No.3 represent these soils.

Sector D: These soils are located at the downstream of El-Shaboura canal, so it is subjected to irrigation with polluted water (domestic dumping). Soil profile No. 4 represent these soils.

Sector E: The soils of this sector are subjected to irrigation with combination of El-Shaboura canal water and either Mostorod collector wastewater (Profiles No. 5, 6, 7) or Shebin El-Qanater collector wastewater drain (Profiles No. 8, 9, 10).

Sector F: These soils are subjected to irrigation from El-Boulaqeyah canal water and Laza agricultural drain wastewater (profiles, No. 11, 12, 13, 14 and 15).

Sector G: The source of irrigation water for the soils of this sector is heavily polluted wastewater of Mostorod collector drain, which receives wastewater from steel, staining, paper, and detergents factories (Profile No. 16).

Soil analyses:

Disturbed soil samples were collected from different layers of the studied soil profiles. These samples were air-dried, crushed, ground and sieved through 2 mm sieve. Fractions below 2 mm were maintained and stored in plastic bottles for analyses. Soil physical and chemical characteristics were discussed else- where [8]. Soil particle size distribution was determined according to Pipette method [9]. Separation of grain size fractions was carried out according to Shuman [10]. A sample from each fraction was collected and stored for heavy metals determination. Heavy metals content (Zn, Cu, Co, and Cd) either in the whole soil sample or in the different soil fractions were extracted by wet digestion of 1.0 g. sample with $HF/HClO_4$ [11].. Heavy metals in soils were measured by Ion coupled plasma (ICP).

The enrichment factors were be calculated according to Davies [3] by the determination of metal content for each fraction (M_f asμg/g) which is multiplied by the fraction % in soil (F %) to contribution of each fraction (C_f) as shown in equation 1.

$$C_f = (F\ \%) \times M_f \qquad (1)$$

Then the sum of C_f 's for Clay, silt and sand gives us the total content bound to the soil fractions (S).

fig. (1)Location of water samples and soils profiles

The resulting metal content contribution of each fraction (P)was expressed as a percentage of the total (S) as shown in equation 2.

$$P = C_f/S \qquad (2)$$

When P was divided by the percentage grade size of each fraction (F) the enrichment factor (EF) for each fraction in tested layers in the selected soil profiles was obtained.

RESULTS AND DISCUSSION :

Data in Table (1) show that in general, the soils subjected to irrigation with wastewater have the highest values of clay and silt content in surface or subsurface layers compared with uncultivated control soil.

Table (1) Soil fractions % and total content of Zn, Cu, Co and Cd in investigated soils

Profile groups	Soil depth (cm)	Grain size fractions %			Total heavy metals (μg/g)			
		clay	silt	sand	Zn	Cu	Co	Cd
A	0-15	45.80	31.50	20.00	83.9	62.2	13.2	1.6
	15-30	43.80	35.50	19.50	82.9	63.4	14.1	1.6
B	0-15	34.78	32.00	28.20	155	60.0	14.5	1.2
	15-30	47.03	26.82	32.50	168	63.5	14.0	1.3
C	0-15	55.60	23.45	18.00	255	197	62.5	8.5
	15-30	62.18	29.32	6.20	214	142	49.5	2.6
D	0-15	70.85	22.95	3.50	199	106	42.0	13.0
	15-30	72.58	21.70	5.10	264	105	37.0	4.0
E	0-15	64.72	28.93	20.70	177	112	34.3	6.5
	15-30	49.13	24.61	21.30	136	104	31.0	4.0
F	0-15	52.30	28.29	15.70	125	98	25.8	1.7
	15-30	49.61	31.63	16.45	83	87	23.2	1.1
G	0-15	33.15	33.73	26.80	561	136	47.0	7.9
	15-30	34.70	31.50	25.20	381	103	36.0	7.1

As shown in Table (1), total Zn content in the studied soils ranges between 82.9 and 561 μg/g. Uncultivated control soils (A) showed the lowest values (82.9-86.1 μg/g) while, in cultivated control soils (B) these values ranged between 155 and 173.0. The normal level of total Zn in alluvial soils of Egypt ranges between 60 and 100 μg/g with an average of 80.0 μg/g (12). The highest level of total Zn (561 μg/g) was observed in the surface layer of soils of group G (subjected to irrigation with wastewater of Mostorod collector drain). It could be noticed that it is 7 times higher than the uncultivated control soil (A) and 2.5 folds compared to the cultivated control soil B (subjected to irrigation with Nile water. A significant increase in total Zn could be observed in the surface layers of soils of groups G, C and D due to the irrigation with wastewater of Mostorod and Shebin El-Qanater

collectors and polluted water of El-Shaboura canal. This may be attributed to the association of Zn with the relatively high content of organic matter in the surface layers of these soils. Analysis of wastewater of six water bodies in the studied area are discussed in details in another work [7].

Total amount of Cu in uncultivated control soil (A) ranges between 61.8 and 63.4 µg/g while in the cultivated control soil B (irrigated with Nile water) ranges between 56.3-63.5 µg/g (Table 1). The highest values are those of the soils subjected to irrigation with different wastewater's (soils of groups C,D,E and G). The total amounts of Cu in the surface layers of these soils are 197.3, 106.8, 112.4 and 135.5 µg/g, respectively. As shown in Table (1) the amounts of total Cu decrease with soil depth. Similar results were reported for the same area [13-15] reported that both total and avai-lable Cu increased in the surface layer of the alluvial soils at El-Saff area due to industrial activities.

The amount of total Co in uncultivated control soil ranges between 13.12 and 14.10 µg/g while, in cultivated control soils (subjected to irrigation with Nile water) it ranges between 14.0 and 15.0 µg/g. El-Sayad and Hegazy [16] reported that the average value of total Co in some alluvial soils of Egypt reported to be 18.05 µg/g, while, Rashad et al [17] reported that this value in the normal alluvial soils of Nile Delta ranged between 3.7-5.5 µg/g with an average of 4.7. The highest values of total Co are obtained in the surface layers of soils of groups C, G (subjected to irrigation with wastewater of Shebin El-Qanater and Mostorod collector drains) and D (irrigated with polluted water of EL-Shaboura canal). These values are 62.5, 47.0 and 42.0 µ/g, which are higher than those of uncultivated control soil (A) by 4.7, 3.6 and 3.2 folds respectively.

Total amounts of Cd in investigated soils are given in Table (1). The amounts of Cd in uncultivated control soil (A) range between 1.5 and 1.6 µg/g with an average of 1.55 µg/g while, in cultivated control soil B (irrigated with Nile water) ranged between 1.2-15 µg/g. Similar data were reported for the normal alluvial soils of Egypt to be from 1.0-1.4 µg/g with an average of 1.2 µg/g [17]. Many authors reported that Cd contents of soil in the non-polluted areas are usually below 1.0 µg/g [18-20]. The highest values of total Cd are observed in the surface layers of soil groups, D and G as they reached 8.5, 13.0 and 7.9 µg/g, respectively. While total Cd in deepest layers of these soils was 0.80, 1.90 and 3.40 ppm, respec-tively. Generally, total contents of Cd in the surface layers of these soils (C,D and G) are higher than those of control soil (A) by 5.3, 8.2 4.9 times respectively. These findings are due to the irrigation with high Cd water levels from Shebin El-Qanater collector, Mostorod collector drain and El-Shaboura canal.

Distribution of tested metals between soil different grain size fractions:

The amounts of Zn, Cu, Co and Cd in clay, silt and sand fractions in surface and subsurface soil samples are shown in Table (2). Data indicate that the average amounts of heavy metals in different fractions are proportional to the total content of these metals in the investigated soils. The average amounts

in the surface 15 cm ranged from 15 to 686; from 8 to 210; from 2.5 to78.21; from 0.22 to 13.50 μg/g for Zn, Cu, Co and Cd, respectively. In all metals, the highest values were found in the clay fraction, while the lowest values were found in the sand fraction.

In uncultivated control soil (A) the concentrations of Zn in clay, silt and sand fractions ranged between 85.0-88.0 μg/g; 96.0 -101.0 μg/g and 57.0-60.0 μg/g, while in cultivated control soil B (irrigated with Nile water) are 219.0-234.0; 135.0-153.0 μg/g and 48.0-86.0 μg/g respectively (Table 2). The highest amounts of Zn are found in the surface layers of groups C and G (subjected to irrigation with wastewater of Shebin El-Qanater and Mostorod collector drains, respectively), where it reaches 354 - 563 μg/g; 158-418 μg/g and 124 - 636 μg/g for clay, silt and sand fractions, respectively.

Generally, clay fraction has the highest percentage of total Zn in soil (41.4-43.1%) followed by silt fraction (34.4-35.1%) and sand fraction (21.8-24.2%). In case of group G, sand fraction has the highest percentage of total Zn (32.0-39.3%), while clay fraction has 33.9-34.8%.

Both uncultivated (A) and cultivated (B) control soils, have an average mean of 80, 71.5 and 30.5 μg/g Cu in the clay, silt and sand fractions, respectively. The clay fraction contributes the highest ratio of total Cu (49.5-59%) followed by silt fraction (34.0-41.4%) and sand (3.9-10.8). Data indicate that irrigation with polluted wastewater tended to cause a noticeable increase of Cu in both clay and silt fractions (Soils of group C, D, E and G). The highest Cu values were observed in the surface layer of soils groups C and G. The total Cu content in the clay fraction was as high as 165 - 210 μg/g with an average of 87.5 μg/g (about 48.5% of total), silt fraction has 137-180 μg/g (mean=158.5 μg/g) about 27.5% and sand fraction has 141-155 μg/g (mean= 148.0 μg/g) about 20% othe tot.

Data for different soil fractions content for Co, in both uncultivated (A) and cultivated (B) control soils, ranged from 18.0-20.5 μg/g; 8.8-16.7 μg/g and 2.5-3.2 μg/g in the clay, silt and sand fractions, respectively. Concerning inves-tigated soils subjected to irrigation with different wastewater, it seems that both soils of groups C and G have the highest values where the three fractions, clay silt and sand in the surface layers have 72.50-78.21, 49.10-77.8 and 3.50-12.5 ppm, respectively. It is clear that the enrich-ment of Co in the clay fraction is about 10 times the sand fraction. Also, data indicated that clay fraction in these soils contributes 51.9 to 52.1% of the total Co, while silt fraction has 39.8 to 40.0 % and sand fraction has 8.1%.

Distribution of Cd between different grain size fractions of the investigated soils ranged between 1.8-2.4, 0.73-1.55 and 0.35-0.70 μg/g for clay, silt and sand fractions In both uncultivated control soil (A) and cultivated control soil (B), res-pectively. The enrichment of Cd in the clay fraction was about 4 times higher than in sand fraction. The highest amounts of Cd content were found in the three fractions of the surface layers of soils of groups C, D, E and G. Clay fraction exhibited the highest values (8.8-13.5 μg/g) followed by silt fraction (5.4-12.1 μg/g) and sand fraction

(0.72-2.80 μg/g). The enrichment of Cd in the clay fraction of these soils is about 7 times the sand fraction. Clay fraction of the surface layers of these soils has the highest ratio from 42.8 to 69.0% of the total Cd with an average of 52.1%, silt fraction has 27.6 to 54.0% with an average of 39.5 % and sand fraction has 3.4 to 14.0% with an average of 8.4%.

Enrichment factor of heavy metals in different grain size fractions:

The enrichment factor of heavy metals in different soil fractions of different soil layers as affected by the irrigation with different polluted wastewater were calculated and presented in Table (3). Presented data indicate that the clay fraction is enriched with most heavy metals compared with other soil particles, particularly in surface layers (0-15 cm). It is clear, that Zn and Cu tend to accumulate in both clay and silt as shown in Table (3). It is worth to mention that the increase of EF- values with soil depth, in some cases, may suggest that some heavy metals in the contaminated soils are leach-able and mobile to some extent through the soil profile due to the presence of organic complexes usually found in wastewater effluent. These dissolved organic compounds act as chelating agents and protect metals from the ordinary precipitation and fixation in surface layer of arid soils. Fig (2) shows the enrichment factor for each soil fraction in surface and subsurface layers of different soil profiles as affected by wastewater application. Also, EF as a ratio may allow the comparison between different heavy metals enrichment under different soil conditions. It is clear that the highest EF ratios in the clay fraction were mainly with Cd and Co which may reflects the industrial waste-water impact on the soil. Moreover, EF values were higher in silt fraction of surface layers than in subsurface layers. In case of sand fraction, Cu and Zn EF values were the highest always compared to other studied metals.

CONCLUSION :

The enrichment factor is a good indicator for potential metal accumulation in soil profile as well as in soil fractions. Also, EF as a ratio may allow the comparison between different heavy metals enrichment under different soil conditions.

Table (2) Distribution of tested metals (μg/g fraction) between various particle size fractions.

Group	Depth cm	fraction	Zn	Cu	Co	Cd
A	0-15	Clay	85.80	72	18.80	2.40
		Silt	95.56	64	9.58	0.73
		Sand	56.60	30	3.20	0.62
	15-30	Clay	88.00	75	20.50	2.40
		Silt	101.00	61	8.80	0.88
		Sand	60.00	30	3.00	0.70
B	0-15	Clay	219.00	86	19.40	1.80
		Silt	153.00	69	15.50	1.15
		Sand	48.30	31	2.50	0.35
	15-30	Clay	234.50	86	18.00	2.00
		Silt	135.70	92	16.70	1.55
		Sand	86.70	31	3.00	0.55
C	0-15	Clay	354.50	210	72.50	8.50
		Silt	157.90	180	77.80	6.90
		Sand	124.00	155	12.50	1.78
	15-30	Clay	230.20	100	32.10	1.95
		Silt	160.20	170	25.20	1.80
		Sand	183.80	82	6.10	0.40
D	0-15	Clay	213.20	110	43.72	12.82
		Silt	142.00	91	36.80	8.62
		Sand	160.50	66	10.50	2.80
	15-30	Clay	147.10	100	25.92	1.68
		Silt	102.60	85	22.10	1.50
		Sand	89.20	50	4.80	0.50
E	0-15	Clay	146.20	106	42.10	9.60
		Silt	158.57	103	53.00	12.10
		Sand	120.55	98	8.00	0.72
	15-30	Clay	109.23	98	31.30	1.40
		Silt	133.65	94	36.20	0.59
		Sand	157.08	52	5.50	0.30
F	0-15	Clay	89.00	116	28.46	0.98
		Silt	88.00	114	23.31	0.86
		Sand	26.55	10	4.80	0.30
	15-30	Clay	90.00	87	21.84	0.80
		Silt	79.65	72	18.68	0.55
		Sand	15.50	8	2.80	0.22
G	0-15	Clay	563.00	165	78.21	13.50
		Silt	418.00	137	49.10	5.40
		Sand	636.00	141	3.50	0.67
	15-30	Clay	184.50	96	36.50	5.40
		Silt	186.00	131	35.00	2.90
		Sand	173.00	65	4.00	0.68

Table (3) Enrichment factors for selected heavy metals in different soil particles for surface and subsurface soil layers.

Profile group	Layers	Zn			Cu			Co			Cd		
		Cl.	Si.	Sa.	Cl.	Si.	Sa.	Cl.	Si.	Sa.	Cl.	Si.	Sa.
A	Surface	1.05	1.16	0.69	1.20	1.08	0.50	1.51	0.79	0.26	1.64	0.50	0.42
	Sub-surface	1.01	1.11	0.73	1.24	0.98	0.50	1.50	0.80	0.26	1.68	0.48	0.42
B	Surface	1.56	1.09	0.33	1.39	1.00	0.50	1.55	1.24	0.20	1.62	1.04	0.31
	Sub-surface	1.43	1.10	0.43	1.19	1.36	0.40	1.80	0.88	0.29	1.61	1.03	0.33
C	Surface	1.37	0.61	0.48	1.11	0.95	0.82	1.19	1.27	0.20	1.25	1.01	0.26
	Sub-surface	1.10	0.82	0.93	0.88	1.05	1.79	1.12	1.00	0.20	1.23	0.71	0.29
D	Surface	1.15	0.84	0.77	1.07	0.89	0.64	1.09	0.92	0.26	1.14	0.77	0.48
	Sub-surface	1.10	0.76	0.62	1.07	0.89	0.54	1.07	0.94	0.24	1.13	0.76	0.21
E	Surface	1.23	1.00	0.70	1.30	1.09	0.50	1.14	1.43	0.22	1.64	1.04	0.11
	Sub-surface	1.09	1.19	0.67	1.04	1.15	0.79	1.18	1.44	0.23	1.28	1.42	0.01
F	Surface	1.32	0.80	0.54	1.28	1.27	0.13	1.35	1.00	0.30	1.45	0.98	0.17
	Sub-surface	1.41	0.87	0.40	1.28	1.15	0.16	1.33	1.08	0.23	1.68	0.64	0.44
G	Surface	1.04	0.77	0.17	1.11	0.93	0.96	1.80	1.12	0.08	2.07	0.83	0.10
	Sub-surface	0.42	0.59	1.97	1.01	1.40	0.66	1.92	0.97	0.08	1.99	0.88	0.09

Fig(2) Enrichment factors of metals in surface layer

Enrichment factors of metals in sub- surface layer

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معامل إثراء المعادن الثقيلة فى حبيبات التربة المختلفة
كمؤشر على تلوث التربة

ف. هـ. ربيع ، م.ف. عبد الصبور ، أ.ت. مصطفى ، س.أ. حسن

تم اختيار المنطقة الصناعية شمال القاهرة الكبرى لدراسة تأثير النشاط الصناعى المكثف على مواصفات التربة ومحتواها من المعادن الثقيلة.

وقد قسمت منطقة الدراسة الى ستة قطاعات طبقا لمصدر مياه الرى واحتمالات التلوث. تم تجديد ١٦ موقع واخذت عينات من التربة وتمت عملية التجفيف فى الهواء وتم تقسيمها الى مجموعات طبقا لحجم الحبيبات. وقد قدرت كميات المعادن الثقيلة مثل الزنك والنحاس والكوبلت والكادميوم فى عينات التربة باستخدام طريق ICP

وقد اسفرت النتائج عن زيادة معامل EF نسبة عمق التربة فى بعض الحالات ربما يفترض ان بعض المعادن الثقيلة فى التربة الملوثة تكون متحركة ومتفاعلة خلال طبقات التربة بسبب وجود المركبات العضوية فى مياه الصرف

وهذه المركبات العضوية الذائبة تساعد المعادن الثقيلة على عدم الترسيب والثبات فى التربة الفاصلة . ووجد أن أعلى نسبةُ EF فى عينات الطمى تكون أساسا بعنصر الكادميوم الذى يفسر التأثير الصناعى على التربة. وفيما يختص بالطبقات الأخرى فقد لوحظ اختلاف تجمع معادن الزنك والنحاس حسب عمق ونوع التربة