

Dynamics of Cadmium and Lead in Some Soils of Chittagong, Bangladesh

M. G. Kibria, M. J. Ahammad, M. Islam and K. T. Osman
Department of Soil Science, University of Chittagong, Chittagong 4331, Bangladesh

Abstract: Total, 1N NH₄OAc and 0.1N HCl extractable Cd and Pb were determined in soils of four sites of Chittagong City contaminated by city sewage (Site-I), tannery (Site- II), fertilizer factory (Site- III) and cement factory (Site- IV) and collected on profile basis and distance from the source points. Total Cd contents of soils in different sites ranged from 0 to 2.48 mg kg⁻¹ while the values for Pb varied from 7.14 to 54.31 mg kg⁻¹. Ammonium acetate and 0.1 N HCl extractable Cd in the surface soils were in the range between 0.01 and 0.62 mg kg⁻¹ and between 0.03 and 1.66 mg kg⁻¹, respectively. Lead contents in surface soils extracted with 1N NH₄OAc and 0.1N HCl varied from 0.18 to 2.72 mg kg⁻¹ and 2.00 to 50.51 mg kg⁻¹ respectively. A close relationship between the total amounts of Cd and Pb and their extractable amounts with 1N NH₄OAc and 0.1N HCl was found. The concentrations of total and extractable Cd and Pb were inversely correlated with soil pH. Strong positive correlations were observed for Cd and Pb with CEC and organic matter contents of soil.

Keywords: Dynamics, cadmium, lead, city sewage, tannery.

I. Introduction

Soil pollution by heavy metal is a major environmental problem facing the modern world [1, 2, and 3]. The metal content in soil is a sum of metals originating from natural processes and human activity. It is estimated that the contribution of the metals from anthropogenic sources in soil is higher than that from natural ones [4]. In recent years, one potential problem that has been recognized in connection with the disposal of sewage waste water to agricultural land is the accumulation of heavy metals in the soil and their uptake by plants [5]. Wastewater irrigation provides water, N, P and organic matter to the soils [6], but there is a concern about the accumulation of potentially toxic elements such as Cd, and Pb from both domestic and industrial sources [5]. Significant increases in soil metal content are found in areas of high industrial activity where accumulation may be several times higher than the average content in non-contaminated areas [7]. The vicinities of many industrial complexes are particularly liable to such high levels of toxic heavy metals that are derived from the discharge of poorly treated liquid effluents to land [8, 9]. There are abundant data on enrichment of heavy metals in soil around strong industrial emission sources [10, 11, 12, and 13].

The distribution of heavy metals in soil profiles, as well as their availability, is controlled by the nature of the metal species, their interaction with soil colloids, and soil characteristics such as soil pH, clay, organic matter and moisture content [14, 15]. It is important to study the chemical forms, mobility and distribution of these toxic metals in contaminated soils since these metals may eventually be translocated to plant tissues [16, 17] and may pose a threat to human health. Assessment of the metal distribution within soil profiles provide a basis for establishing long-term wide spread predictable changes in soil and ground water quality. Food chain contamination by heavy metals has become a burning issue in recent years because of their potential accumulation in biosystems through contaminated soil, water and air. The basis for general concern about heavy metals getting into the human food chain is the gradual spread of heavy metals into agricultural soils through industrial and urban activity [18].

A considerable works have been done on the heavy metal contamination of soil and their uptake by plants by anthropogenic sources such as industrial operation and urbanization in most developed countries. However, there is paucity of information from developing countries like Bangladesh, and more so, relatively few studies have been reported on the heavy metal status of soils around Dhaka city [19, 20, 21]. Hence, the assessment of metal contamination is of vital importance in industrial areas of Bangladesh. Chittagong city is the second largest city and the main industrial zone of Bangladesh with an area of 209.66 square kilometres and a population of about 3 millions [22]. A large number of industrial operations including plastic, textile, tannery, chemical industries, paints and dye manufacturing, oil refinery and some of the known industries like TSP Complex Limited, Heidelberg Cement Bangladesh Limited at Potenga etc. are located in the municipal area. With increased disposal of urban and industrial sewage waste on agricultural lands of Chittagong, there is the need to understand further the mobility of heavy metals in soil, especially those of Cd and Pb. With a view to understanding the heavy metal dynamics in soil the present study was carried out with the following objectives:

1. To assess the level of contamination and vertical and horizontal distribution of Cd and Pb in soils around Chittagong Municipal area contaminated by city sewage, tannery, fertilizer and cement factory.

2. To evaluate the relationships among total and extractable Cd and Pb and soil properties.

II. Materials and Methods

2.1. Sampling Sites of the Study Area

Chittagong city is situated within 22°- 14' and 22°- 27' N Latitude and between 91°- 46' and 91°-53' E Longitude. Four sampling sites were selected in the study area. The sites were (i) Site-I. City sewage contaminated site to which city sewage has been disposed of for about 50 years (ii) Site-II. Tannery contaminated site (iii) Site-III. Fertilizer factory contaminated site and (iv). Site-IV. Cement factory contaminated site. City sewage (22°-22'-89" N and 91°- 50'-16" E) and tannery (22°-24'-10" N and 91°- 49'- 14" E) contaminated sites are located at Haji para and Koolgaon, respectively in the low lying areas of northern part of the Chittagong city. On the contrary, the fertilizer factory (22°-16'-34" N and 91°- 47'- 62" E) and cement factory (22°-16'-71" N and 91°- 47'- 80" E) contaminated sites are located at Potenga in the high lying areas of southern part of the city. Location and soil sampling sites of the study area are shown in Fig. 1.

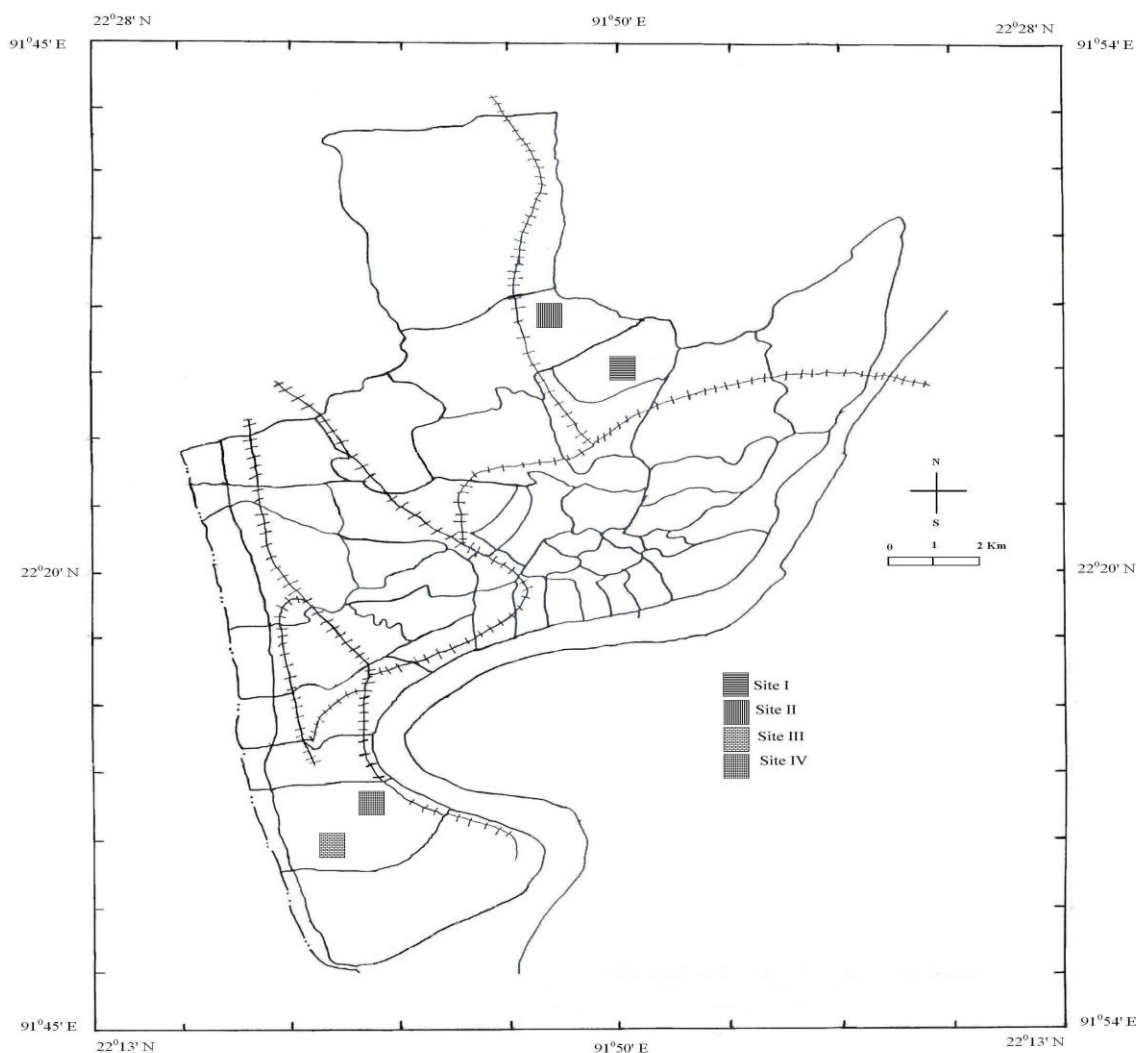


Fig. 1 Location and soil sampling sites of the study area

2.2. Collection of Soil Samples

Soil samples were collected on profile basis. Three profiles were dug in each site. One profile was dug in the vicinity of the source point (Profile-1) and another two were 0.5 km (Profile-2) and 1.0 km (Profile-3) away respectively from the source point in order to examine how the total contents of Cd and Pb and their mobility change with increasing distance from the source of contamination. Soil samples were collected from dug pits from different depths down to a maximum depth of 82 to 135 cm. Composite samples of each horizon were taken with four to five sub samples from the walls of a pit. Soil sampling was initiated from the lower most horizons to avoid contamination. The samples were air dried and sieved to < 2 mm and were preserved for physical and chemical analysis.

2. 3. Soil Analysis

The particle size distribution was determined by hydrometer method of Day [23]. Soil pH was measured in a 1:2.5 soil/water suspension with glass electrode pH meter. The potassium dichromate wet-oxidation method of Jackson [24] was used for the determination of organic carbon followed by multiplying the values with 1.724 to calculate the organic matter contents. The cation exchange capacity was determined by saturation with 1N NH₄OAc at pH 7.0 [24]. The soil samples were digested with aqua regia [24] on a sand bath for the determination of total Cd, Pb, Zn, Fe, Mn, P and K. In order to determine the mobility of Cd and Pb in the soils, the extractable amounts of the metals were determined after extraction with 1N NH₄OAc and 0.1 N HCl [25]. Total nitrogen was determined by micro - Kjeldahl method as described by Jackson [24]. Phosphorus was determined by vanadomolybdo phosphoric yellow color method in nitric acid system according to Jackson [24]. Potassium was measured by flame photometer and Fe, Mn, Zn, Cd and Pb were determined by atomic absorption spectrophotometer (Varian spectra AA-220).

2. 4. Data Analysis

The significance of differences between the means of the treatments was evaluated by one way analysis of variance followed by Duncan's Multiple Range Test at the significance level of 5%. Pearson's correlation coefficient was estimated to test the relations among metal contents in soil and soil properties. The statistical software Excel [26] and SPSS version 12 [27] were used in the analysis.

III. Results and Discussion

3. 1. General Soil Properties

Soil pH ranged from 4.9 to 5.4, 6.2 to 8.3, 5.4 to 6.8 and 6.3 to 7.2 in site- I, II, III and IV respectively. The percent sand, silt and clay content of the surface soils among the contaminated sites were in the range of 5 to 60, 14 to 41 and 22 to 57, respectively. Texture of the surface soils varied from sandy clay loam to clay. Organic matter varied from 0.14 (lowest horizon of profile 2) to 3.51% (surface horizon of profile1) in site I. The organic matter contents were 0.21- 1.95 %, 0.08 - 2.36 %, and 0.08- 1.93 % in site II, III and IV respectively. The surface horizon contained the highest amount of organic matter in each site. The organic matter content decreased with depth. Among the sites, site I contained the highest organic matter. The surface horizons of three profiles of site I and the profile adjacent to the source point in other three sites contained the organic matter level higher than that normally present in soils of Bangladesh. According to BARC [28], the normal range of organic matter is <1.0 -1.7 % in soils of Bangladesh.

The cation exchange capacity in site-I was in the range of 3.13 - 13.35 cmol kg⁻¹ soil while these values were 3.11-9.31, 4.06 -12.51 and 4.05-11.97 cmol kg⁻¹ soil in site II, III and IV respectively. The cation exchange capacity was largely determined by the organic matter and clay contents. So the highest CEC was found in soil rich in organic matter and clay. However, the CEC values did not show any definite trend of variation with depth.

3. 2. Total Cadmium and Lead

Total Cd and Pb contents varied widely in the soil profiles (Table 1). Total Cd ranged from 0.14 to 2.48 and Pb from 8.24 to 54.31 mg kg⁻¹ in the soil profiles of site I. In comparison to this site, the total Cd and Pb concentrations were lower in profiles of site II, III and IV respectively with the ranges 0.06 - 0.24, 0.06 - 0.27 and 0.07-1.03 mg kg⁻¹ for Cd and 7.98 - 23.84, 7.14 - 40.29 and 8.52 - 33.16 mg kg⁻¹ for Pb respectively. The total Cd concentration in surface soils in profile 1 of site I and IV was many folds higher than the worldwide mean Cd concentration of 0.53 mg kg⁻¹ in surface soils [17], threshold levels of ≤ 0.6 mg kg⁻¹ in natural background soil as defined by China [29], and the guidelines value of total soil Cd concentration by the Swiss Ordinance of 0.8 mg kg⁻¹ [30]. The values are also above the Canadian guidelines for maximum allowable concentrations in agricultural soils (1.4 mg kg⁻¹) [31]. The total Pb concentration of surface soils in profile 1 of all the sites was also found above the normal range for soils (5-10 mg kg⁻¹) quoted by Ashwathanarayana [32]. In a study, Kashem and Singh [20] observed that the soils of several industrial areas around Dhaka city were contaminated by Cd and Pb in the ranges of 0.11-1.80 mg kg⁻¹ and 17.1- 89.6 mg kg⁻¹, respectively. The Cd and Pb concentration of 2.60 mg kg⁻¹ and 136 mg kg⁻¹ in contaminated soils from Tejgaon industrial area was reported by Chamon *et al.* [33].

The high contents of these metals especially of Cd in the surface layer of the profile 1 of site I in comparison with profile 2 and 3 of the same site indicate their contamination via the city sewage received by this soil over about 50 years. This is in conformity with Chen *et al.* [34] who investigated the influence of previous irrigation of sewage water from industry and daily life on the concentrations and chemical forms of heavy metals in soils at Heqin Town, Shanghai, China. They found that the total amounts of Cd in the soil at distances of 2, 10 and 20 m from an abandoned open canal exceeded the permissible value set by the Chinese

Table 1. Cd and Pb concentration (mg kg⁻¹) in soils of different contaminated sites

Site No	Profile No	Depth (cm)	Total		NH ₄ OAc Extractable		0.1N HCl Extractable		
			Cd	Pb	Cd	Pb	Cd	Pb	
Site I	1	0 - 10	2.48	54.31	0.62	2.72	1.66	50.51	
		10 - 25	2.05	32.49	0.49	1.29	1.33	27.94	
		25 - 53	1.16	23.44	0.26	0.82	0.63	18.76	
		53 - 85	0.49	14.36	0.10	0.43	0.27	12.06	
	2	0 - 15	1.08	27.32	0.32	0.82	0.84	20.49	
		15 - 32	0.56	20.86	0.11	0.52	0.32	14.19	
		32 - 49	0.33	16.06	0.05	0.32	0.15	9.79	
		49 - 82	0.26	8.24	0.05	0.13	0.15	5.93	
	3	0 - 13	0.14	42.76	0.03	0.85	0.07	22.24	
		13 - 25	0	25.53	0	0.26	0	10.73	
		25 - 55	0	21.36	0	0.16	0	7.26	
		55 - 90	0	12.97	0	0.11	0	3.89	
Site II	1	0 - 15	0.24	23.84	0.03	0.95	0.09	5.96	
		15 - 50	0.09	16.77	0.01	0.50	0.03	3.69	
		50 - 90	0.07	11.55	0.01	0.29	0.02	2.31	
	2	0 - 18	0.06	19.36	0.01	0.19	0.03	3.68	
		18 - 55	0	13.29	0	0	0	2.26	
		55 - 95	0	10.07	0	0	0	1.41	
	3	0 - 16	0.07	15.39	0.01	0.15	0.03	2.00	
		16 - 52	0	11.59	0	0	0	2.08	
		52 - 88	0	7.98	0	0	0	1.19	
	Site III	1	0 - 15	0.27	40.29	0.06	1.21	0.16	7.25
			15 - 75	0.11	15.44	0.02	0.31	0.06	2.16
			75 - 120	0.07	13.07	0.01	0.13	0.04	1.31
2		0 - 20	0.08	34.95	0.01	0.70	0.04	4.54	
		20 - 90	0	13.68	0	0.13	0	1.23	
		90 - 135	0	12.16	0	0	0	1.22	
3		0 - 18	0.06	18.23	0.01	0.18	0.03	2.37	
		18 - 80	0	10.33	0	0.05	0	1.24	
		80 - 130	0	7.14	0	0	0	1.00	
Site IV		1	0 - 20	1.03	33.16	0.18	0.83	0.49	12.27
			20 - 40	0.88	20.83	0.13	0.42	0.38	5.21
			40 - 80	0.42	21.44	0.07	0.22	0.19	7.07
	80 - 120		0.29	15.12	0.04	0.15	0.13	4.99	
	2	0 - 15	0.17	28.49	0.02	0.28	0.07	7.41	
		15 - 35	0	18.44	0	0.09	0	4.42	
		35 - 70	0	14.32	0	0	0	2.86	
		70 - 125	0	13.06	0	0	0	3.66	
	3	0 - 17	0.07	17.68	0.01	0.18	0.03	3.18	
		17 - 42	0	12.84	0	0	0	2.31	
		42 - 85	0	14.68	0	0	0	2.94	
		85 - 122	0	8.52	0	0	0	1.45	

Environmental Quality Standard (GB 15618-1995) and the contamination showed a trend of decreasing with increasing distance from the canal. Lu *et al.* [35] also found a significant influence of distance on the concentration of soil heavy metals in a sewage irrigated agricultural area within 25 m of a canal in which the sewage flowed. The highest concentrations of Cd and Pb found in city sewage area soils (Site I) among the sites under study might be due to liquid wastes, flocculated sludge and other solids with excessive heavy metals coming from anthropogenic activities in Chittagong city.

The contribution of the tannery industry with heavy metals in the present study was evident from the decreasing concentrations of Cd and Pb found with increasing distance from the disposal points of wastes and effluents of site II (Table 1). Significant contribution of tannery industry towards the accumulation of heavy metals such as Cr, Cd, Pb, Zn, Cu in soils was reported by Nuruzzaman *et al.*, [36] and Kashem and Singh [20]. Kashem and Singh [20] found the concentrations of Cd, Pb, Zn, Mn and Cu to decrease with distance from the disposal points of tannery industries located at Dhaka Municipality area. They reported the mean Cd and Pb concentrations of surface soils of 1.26 and 68.10 mg kg⁻¹ soil, respectively in the tannery area, about 2 km away from the main disposal points of wastes and effluents.

Higher contents of Cd and Pb in source point and in surface soil were observed in the fertilizer factory site (Site III). The accumulation of Cd and Pb by the fertilizer plant might be through their emission into the atmosphere, or by depositing the raw material or byproduct nearby the plant or by the overflow of tailing discharges in water resources. The specific pollutants of Cd, Sr, Th, Ce, U, Sm, Mo, Ni, V and Zn emitted into the air by fertilizer industry were reported by Oprea and Pincovschi [37].

The highest Cd and Pb concentrations in surface soils in cement industry site (Site IV) of this study were recorded in profile 1 i. e. adjacent to the Cement Plant (Table 1). This could be attributed to the kiln dust, a rich source of heavy metals including Cd and Pb, released during the processing and production of cement [38, 39]. Cadmium concentration in surface soil of profile 1 in this study was about 15 times higher than that of profile 3 (1 km away from the source point). Oluokun *et al.* [40] reported the elevated Cd and Pb levels ranging from 18 to 35 mg kg⁻¹ and 840-950 mg kg⁻¹ respectively within 5 km radius of the Shegamu Portland Cement Industry in Nigeria.

In site I, considerable amount of Cd moved downwards up to the 53 cm depth in profile 1 but that was not the case with Pb. This may be due to high sand contents and low soil pH in this site [41]. Cadmium release from soil increases substantially when pH drops below 4.5 [42]. This is especially true for sandy soils with low CEC, low acid neutralization capacity, and low ability of the subsoil to sorb Cd ions. Smith [43] showed evidence of such movement in a sandy soil treated with sewage sludge at intervals over a period of 25 years. The distribution of heavy metals in soil profiles is continuously being changed due to both anthropogenic activities and natural turn over in rock-soil-plant systems [21].

In general, the Cd and Pb distribution in the profiles showed a fluent decrease with depth indicating anthropogenic inputs associated with city sewage and industrial operations of the studied areas, and which is in accordance with other studies [44, 45]. The Cd and Pb contents varied significantly between depths within a profile in all the sites with some minor exceptions and the surface layers contained higher amounts of Cd and Pb than the subsurface layers (Table 1).

3. 3. Mobility of Cadmium and Lead

Determination of the mobile forms of heavy metals is important for understanding their migration patterns in the soil and their uptake by plants. The extractable concentrations of metals are considered to represent their bioavailability. According to Sabienė and Brazauskienė [25], the single extractions by 1N NH₄OAc (pH 7.0) and 0.1N HCl of soil were used in this study for assessing mobile forms of Cd and Pb. Metals extracted with 0.1N HCl are thought to represent the mobile fraction of metals and may reflect the bioavailability of metals [46].

Extractable Cd with 1N NH₄OAc in the surface horizon varied from 0.01 mg kg⁻¹ in site III and IV to 0.62 mg kg⁻¹ in site I while 0.1N HCl extractable Cd ranged from 0.03 mg kg⁻¹ to 1.66 mg kg⁻¹ (Table 1). The values of extractable Cd decreased with depth up to zero in most of the profiles. The extractability of Cd by 1N NH₄OAc ranged from 12 to 30 % and by 0.1N HCl ranged from 40 to 78 % of total Cd in the surface horizon. The relative extractability of Cd was the highest in city sewage soil (30% for 1N NH₄OAc and 78 % for 0.1N HCl) followed by fertilizer, cement and tannery soil suggesting the city sewage soil should be of more concern for contaminating agricultural products.

A very small amount of Pb was extracted with 1N NH₄OAc from the surface horizons and the values ranged from 0.15 mg kg⁻¹ in site II to 2.72 mg kg⁻¹ in site I (Table 1). However, 0.1N HCl was able to extract Pb from all soils and it varied from 2 mg kg⁻¹ to 50.51 mg kg⁻¹ in surface horizons, while it decreased with depth to 1.00 mg kg⁻¹ in site III. The relative extractability of Pb by 1N NH₄OAc and 0.1N HCl (% of total Pb) in the surface horizon was in the range of 1 - 5 % and 13- 93 % respectively, while the extractability in all soils decreased with depth as was also the case with NH₄OAc -Cd and HCl-Cd. In general, the extractability was

higher for Cd than for Pb. Cadmium is reportedly more soluble than other metals [47, 48]. Lead was less extractable which might be due to its high affinity to soil organic matter [41, 49, 50].

In the present study, 0.1N HCl extracted the larger amounts of heavy metal than did 1N NH₄OAc. Because it releases heavy metals from many soil ‘pools’, e.g., exchangeable, organically complexed and secondary minerals such as carbonates, oxides and hydroxides of iron and manganese [25].

3. 4. Correlations among total and extractable cadmium and lead and soil properties

A correlation matrix for Cd and Pb and properties of soils of the sites under study was calculated to see whether they were interrelated with each other and the results are presented in Table 2. The extractable amounts of soil metals seem to be of more practical significance for the assessment of soil contamination. However in the present study, there was a close relationship between the total amounts of Cd and Pb and their extractable amounts with 1N NH₄OAc and 0.1N HCl (Table 2). This is in consistent with the results found by Sabienė and Brazauskienė [25]. The concentrations of total and extractable Cd and Pb were inversely correlated with soil pH. The strong negative relationship between extractable metal concentration and soil pH was also observed by many other investigators [51, 52]. Wang *et al.* [52] found that NH₄OAc extractable Cd, Zn and Ni decreased with increasing pH. More than 90 % of the variability of Pb and Zn in the exchangeable fraction was caused by the variability of soil pH [53]. A considerable number of other studies also tried to infer relationships between the mobility of metals and soil properties [54, 55]. In general, soil pH and CEC were found to be important predictors of metal extractability [50], retention and movement in soils [41]. Strong positive correlations of total contents of Cd and Pb with their mobile fractions and each other suggest overloading of geochemical system in the contaminated soils, where anthropogenically added metals remained weakly bound to soil constituents [56].

Table 2 Correlation Coefficients among total and extractable Cd and Pb and soil properties

	Cd _T	Cd NH ₄ OAc	Cd _{0.1 HCl}	Pb _T	Pb NH ₄ OAc	Pb _{0.1 HCl}	Clay	pH	OM	CE C
Cd	.988(**)									
NH ₄ OAc Cd _{0.1}	0.989**	0.999**								
HCl Pb _T	0.709**	0.642**	0.642**							
Pb _{NH₄OAc}	0.840**	0.845**	0.845**	0.872**						
^c Pb _{0.1 HCl}	0.887**	0.908**	0.906**	0.784**	0.889**					
Clay	0.171ns	0.125ns	0.129ns	0.385*	0.209ns	0.089ns				
pH	-0.497**	-0.542**	-0.532**	-	-0.391*	-0.528**	-			
OM	0.576**	0.601**	0.606**	0.403**	0.787**	0.714**	0.094ns			
CEC	0.414**	0.394**	0.402**	0.787**	0.624**	0.456**	0.688**	0.415**		
								-0.258	0.757*	

ns, * and ** represents non significant, significant at p< 0.05 and p< 0.01 , respectively.

The relationships between Cd and soil properties are complex in that they are a result of parent material composition, natural and anthropogenic inputs of Cd as well as intrinsic soil properties which control the speciation and fate of Cd [57]. According to Holmgren *et al.* [58], at below approximately pH 7, Cd concentrations in mineral soils appear to be positively correlated to both soil pH and organic carbon content. Above pH 7, the correlation between Cd and organic carbon is less significant. In the present study, total Cd concentration was correlated strongly and positively with organic matter but total Cd concentration was negatively correlated with pH where measured soil pH was below 7 except for 4 samples.

While the correlation of CEC with soil pH was not significant for the data of the present investigation, strong positive correlations were observed for Cd and Pb with CEC and organic matter (Table 4. 2). In general, the higher the CEC, the greater is the ability of retention of heavy metals [59]. The type and quantity of clay determines the CEC which increases with clay content, particularly when it contains a high proportion of 2:1 lattice type minerals (e.g., montmorillonite). In the present study, clay content did not exhibit any significant relationship with total and extractable metals. However, Chlopecka *et al.* [60] did not observe any consistent relation between heavy metal concentrations and clay content, organic matter, carbonate content, pH and EC.

IV. Conclusion

The results of this study indicate anthropogenic inputs of Cd and Pb in agricultural soils associated with discharge of city sewage, tannery effluents and atmospheric deposition of dust particles from fertilizer and cement factory near the area of investigation around Chittagong Municipality. The level of Cd and Pb contamination of agricultural soils of Chittagong Municipality area is an important concern for food safety and human health. The results of the present investigation highlight the need for establishing soil quality standards for heavy metals to predict human induced soil pollution. For further information about other areas of the Chittagong Municipality, the investigation should be expanded in the near future to enlarge and improve the basis for proposing agricultural soil quality standards at national levels.

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