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Fractionation, Characterization and Speciation of Lead in the Industrially Polluted Soils of Tejgaon Area of Bangladesh and Lead Pollution of Associated Plants and Water

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Abstract

The Tejgaon industrial area is located within the Dhaka Municipality area and about 5 km north of the city centre. At Tejgaon soil pH was around neutrality (mean 6.73) although the minimum value was 5.3 and the maximum 7.6. High buffering capacity resulting from high soil organic matter content (4.87-11.55%) probably influenced soil pH to a neutral value. Electrical conductivity (EC) (25-551 µS), organic matter (OM) (4.87-11.55%) and nitrogen (N) contents were also influenced by the industrial wastes and effluents of the factories. The average Pb concentrations (mg/kg) in different soil fractions were 7.80, 21.85, 56.15 and 130.29 and 11.42, 20.27, 40.33 and 95.08 in water-soluble, NH_4OAc extractable, DTPA (Diethelene-Triamine-Penta-Acetic acid) extractable and total fractions during dry (January) and rainy seasons (September) respectively. Comparing 1st (rainy season) and 2nd (dry season) sampling data, Pb concentrations in different soil fractions were higher in the 2nd sampling data. Concentration of total Pb in soils during 1st sampling were found above the natural background level with few exceptions. Only 4% of total Pb (=20 mg kg^{-1}) samples were in the normal range during wet season. Lead concentration in the group of tolerable level was 58% (>20-≤=100 mg kg^{-1}) and 38% (≥=100 mg kg^{-1}) were found in the group of in excess of tolerable level during 1st sampling. On the other hand 62.5% (≥=100 mg kg^{-1}) were found in the group of in excess of tolerable level (0.01 mg kg^{-1} Pb) during dry season. In case of water samples, total Pb concentration in the rainy season, all samples were found in the group of in excess of tolerable level during 1st sampling. Plant samples were found to contain untolerable total Pb (>20 mg kg^{-1}) during rainy and dry seasons, respectively.

Key words: Speciation, Tejgaon soil, Industrial area, Lead.

Introduction

The presence of heavy metals in soil raises serious concern about the adverse environmental impact as a result of excessive waste and effluent application to agricultural lands. High and excessive accumulation of heavy metals in soil and other media may eventually contaminate both human and animal food chain (He *et al.,* 1992; Iwegbue *et al.*, 2007). Because of this, many countries in the world have established specific guidelines and standards for application of wastes and effluents in agricultural lands. However, these guidelines are lacking in most developing countries. These guidelines, which are generally based on phytotoxic effects and limited to plant uptake studies normally specify the maximum allowable total metal concentration and exhibit considerable variation.

An appreciation of the effect of heavy metals in soil and water on crops can only be attained from precise knowledge of the heavy metal speciation and the response to plant species. In addition, speciation will determine metal mobility and hence potential contamination of ground water following industrial waste application. Although speciation schemes to examine the solid and solution phase of wastes and waste amended soils are subject to analytical limitations. Despite uncertainties resulting from the selectivity of the various extractants and possible problem due to redistribution and re-adsorption phenomena, procedures provide qualitative information on forms, association, bioavailability, their potential effects and guidelines for land application.

Bangladesh has at present about 30,000 large and small industrial units, l. They are discharging their wastes and effluents in the natural systems in most cases without any treatment and thereby cause environmental pollution especially due to heavy metals and organic toxins. The hazardous

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wastes and effluents are generally discharged in low-lying areas along the road sides or in the vicinity of the industrial installations. Besides, the uneducated farmers are randomly using fertilizers and pesticides in agricultural lands. The heavy metals discharged from industries in Bangladesh are cadmium, lead, chromium, mercury, zinc, arsenic and in few cases copper and manganese. The industries like tannery, paper and pulps, textiles, carbides, pharmaceuticals, pesticides, distilleries etc. discharge heavy metals with their effluents and wastes.

Trace metal composition of soil varies widely depending on the sources, industrial waste type (He *et al.,* 1992) and geographical location (Krogmann, 1999). The levels of heavy metals in municipal solid waste have been reported for many countries in the world by several authors (Watanable and Kurihara, 1982; Sridhar and Bammeke, 1986; He *et al.*, 1992; Krogmann, 1999; Koivula *et al.,* 2000). Trace metals can exist in a variety of forms in soil. These forms include (i) water soluble, (ii) exchangeable, (iii) linked to organic substances, (iv) occluded or co-precipitated with oxides, carbonates and phosphates, or other secondary minerals and (v) ions in the crystalline lattices of the primary minerals (Emmerich *et al.,* 1982; Lake *et al.*, 1984; Petruzzelli, 1989; He *et al.*, 1992; Iwegbue *et al.,* 2006b,c). The first three chemical forms are considered to be balanced among themselves; this equilibrium is influenced by pH and the concentrations of metals and ligands. The metals present in these forms are considered to be most available forms for plant nutrition, each successive form representing less availability (Petruzzelli, 1989; Iwegbue *et al.,* 2006b).

The distribution of metals between the specific forms varies widely according to the individual metal and the characteristics of the wastes. These parameters include pH, temperature, oxidation-reduction potential and the presence of complexing ligands.

Total trace metal composition of soil is of little importance in determining its uptake by plants and consequently, in contaminating the food chain since the different forms have different mobilities, bioavailabilities and environmental contamination potential. It is more important to know the distribution of each trace metals in the different forms, rather than just the total content (Lake *et al.,* 1984; Petruzzelli, 1989). Various techniques used to estimate heavy metal distribution in soils, and sewage sludge have been adopted for fractionation of heavy metals in soils. There are two basic techniques. These techniques include (i) physical fractionation based on particle size and density, and (ii) chemical extraction based on selective dissolution of trace metals in different reagents.

In order to asses the speciation or binding forms of heavy metals in solid materials, different analytical procedures involving successive extractions have been developed. These procedures are based on the assumption that the following species of heavy metals exist in soils (Bruemmer *et al.,* 1986):

1. Water-soluble (i.e. in the soil solution)
2. Exchangeable
3. Organically bound
4. Occluded in iron and manganese oxides
5. Definite compounds (e.g. heavy metal carbonates, phosphates, sulphides)
6. Bound in silicate structure (i.e. the residual fraction)

The heavy metals in the soluble fraction can be determined in the saturation extract of soil samples or by extracting soil material with deionized water at a certain ratio (Horak, 1979; Sposito *at al.,* 1982).

In the present research, sequential extraction methods were frequently used to characterize and predict the mobility and bioavailability of heavy metals in soils. The extractants commonly used included deionized water (H_2O), ammonium acetate (NH_4OAc), potassium chloride (KCl), diethylene-triamine-penta-acetic acid (DTPA), and hydrochloric:nitric acid mixture called aqua regia (HCl:HNO3=3:1). Each extractant was targeted on a single form of each metal, for example, H_2O for soluble species, NH4OAc for exchangeable species, DTPA for plant available species and aqua regia for total, noncrystalline form of each metal species (Lake *et al.,* 1984; He *et al.,* 1992). During the course of the present research, soils, plant samples and water samples were collected and analyzed by various independent extractant for characterization of the lead (Pb) species in soils and their subsequent concentration into plants and water.

Material and Methods

Description of the sampling areas

The study area consisted of two distinct areas. One was the lagoon bordering Tejgaon industrial area. This area receives

the drainage residue of the industrial area, Gulshan model town, Niketan Housing Society, Begunbari and other adjoining residential and slum areas including numerous classified/unclassified industries. The other consisted of the canal which receives the inflow from the lagoon and drains into Balu river. In the rainy season the pollution remains minimum because of heavy rainfall; most of the suspended materials, which are not complexed and precipitated with soil, organic matter and other compounds, are flushed out through the canal into the adjoining vast flood zone. The soil and water pollution load increase when the rainy season is over.

The main focus of the study was Tejgaon industrial area which is located within the Dhaka Municipality area and about 5 km north of the city centre. The effluents from many diverse industries in Tejgaon industrial area flow into a lagoon where the residue settle into a thick muck. It overflows through a sluice gate at Rampura into a channel which flows east for about 10 km to Balu river which in turn flows into Sitalakhya river. The whole area is really a flood drainage zone which was originally connected to the Buriganga river on the west of Hazaribagh through a canal passing through Jhikatala, Dhanmondi, Kalabagan, and Hatir

Table I: Identity of different samples with sampling locations. Distances of sampling locations from source of pollution increased according to sample location number

Location	Location Name	Longitude, North	Latitude, East	Samples Type		
1	Begun bari. Lal Tanki	23°45.7	90°24.5	Soil	Water	Grass
2	Boubazar	23°45.8	90°24.6	Soil	Water	Bean leaf
3	Gulshan shooting club (G S C)	23°46.0	90°24.7	Soil	Water	Grass
4	Back side of Arong	23°46.2	90°24.8	Soil	Water	Grass
5	Iron Bridge	23°46.3	90°24.9	Soil	Water	Grass
6	Flood plain	23°46.2	90°25.0	Soil	Water	Grass
7	Flood plain	23°46.1	90°25.1	Soil	-	Grass
8	Rampura Bridge	23°46.1	90°25.4	Soil	Water	Grass
9	Ullan	23°46.1	90°24.9	Soil	Water	Grass
10	Mahanagar Project	23°45.9	90°24.8	Soil	Water	Grass
11	Madhubag	23°45.6	90°24.6	-	Water	-
12	Ha-Meem Garments	23°45.7	90°24.4	Soil	Water	-
13	Nasirabad	23°45.7	90°27.8	-	Water	-
14	Nasirabad ghat	23°45.7	90°27.8	Soil	Water	Water hyacinth
15	Nasirabad	23°45.7	90°27.9	Soil	Water	Water hyacinth
16	Nasirabad	23°45.7	90°28.1	Soil	Water	Bean leaf
17	Guronagar bottala	23°45.7	90°28.4	Soil	Water	Kalmi
18	Guronagar	23°45.6	90°28.7	Soil	Water	Kalmi
19	Balu river	23°45.6	90°28.7	Soil	Water	Water hyacinth
20	Nalchata ghat	23°46.6	90°28.9	Soil	Water	Kalmi
21	Nalchata	23°45.5	90°28.9	Soil	Water	Kalmi
22	Balu river	23°45.55	90°29.0	-	Water	Kalmi
23	Dasher Kandi	23°45.9	90°27.8	Soil	Water	Water hyacinth
24	Trimohini	23°45.6	90°27.5	Soil	Water	Water hyacinth
25	Trimohini	23°45.6	90°27.2	Soil	Water	Water hyacinth
26	Aongargora Meradia	23°46.6	90°26.8	Soil	Water	Water hyacinth
27	Meradia	23°45.7	90°26.5	Soil	Water	Kalmi
28	Aftab nagar	23°45.8	90°26.2		Water	Water hyacinth
29	Banashere	23°45.8	90°25.9		Water	Water hyacinth
30	Rampura Bridge	23°46.0	90°25.6		Water	Water hyacinth

Jheel into Balu river. The canal between Jhikatala and Dhanmondi was closed during the middle of the last century. Overflowing rainwater used to discharge through this canal. Dhanmondi-Kalabagan portion was closed about 20 years ago. Other parts of the vast flood drainage zone are fast being filled up to cater for the development of Dhaka city. The common agricultural crop is rice with substantial amount of various vegetables like chilli, capsicum, carrot, cauliflower, cabbage, corriander, water hyacinth, and tomato. There are abundance of water hyacinth and natural grasses inside the embankment, which are used as fodder for domestic animals.

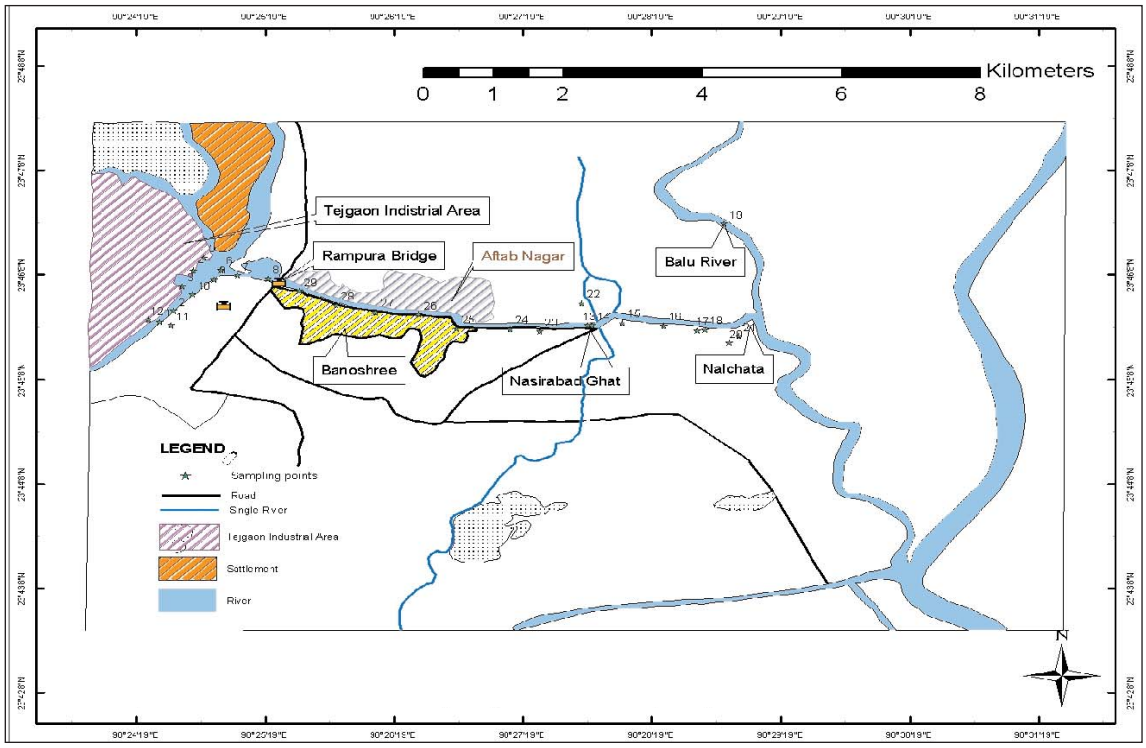

Fig. 1: Location of Sampling Sites During Dry and wet Season

Sample collection and preservation

Soil samples were collected from the study area on two different dates, September, 2006 (wet season) and January, 2007 (dry season). Aerial photographs were downloaded from Google Earth website. The sampling sites were of georeferenced with GPS (Global Positional System) and marked on the map (Figure 1). The location of sampling sites are given in Table I. All samples were collected fresh from the field in required amounts, wrapped in polyethylene bags and transported to laboratory and preserved at +4^{o}C for processing next day.

Soils

The soil samples were collected from 0-15 cm depth. Water soluble, NH_4OAC extractable, DTPA extractable fractions were analyzed and moisture contents of samples were determined on the next day. Rest of the samples were air dried and ground to pass through 2-mm sieve for subsequent physical and chemical analysis.

Metal extraction and determination

i. Water soluble : Soil samples were extracted with deionized water at a soil:water ratio of 1:10 according to Sparks, *et al.* (2001).

ii. NH_4OAc extractable: Soil samples were extracted with NH_4OAc at soil:NH_4OAc solution ratio of 1:10 according to Sparks, *et al.* (2001).

iii. DTPA extractable: Soil samples were extracted with

DTPA solution at soil:DTPA solution ratio of 1:2 according to Sparks, *et al*. (2001).

iv. Total (non-crystalline fraction): Soil samples were digested with aqua regia (HCl:HNO_3) according to Blum, *et al.* (1996).

Plants

All samples were air dried before oven drying at 70°C and ground to powder to pass through a 2-mm sieve for chemical analysis. After digestion the clear solution was adjusted to a volume of 100mL with distilled deionized water.

Digestion with HNO_3-$HClO_4$

One gm of finely ground plant sample was weighed into a 100 mL Kjeldahl flask and digested with 10 mL conc. HNO_3 and 2 mL conc. $HClO_4$.

Water

Water samples (500 mL) were collected from each site in deionized polyethylene bottles fitted with liquid-tight stopper from source water. The samples were immediately acidified with 4 mL of concentrated hydrochloric acid per litre and analyzed within 7 days of collection.

Pb in the extracted solutions, plant and water samples were determined by atomic absorption spectrophotometer in the Centre for Advanced Research on Physical, Chemical, Biological and Pharmaceutical Sciences, Dhaka University of Bangladesh.

Statistical analysis

At Tejgaon soil pH was around neutrality (mean 6.73) although the minimum value was 5.3 and the maximum 7.6. High buffering capacity resulting from high soil organic matter content (4.87-11.55%) probably influenced soil pH to a neutral value. Electrical conductivity (EC) (25-551 μS), organic matter (OM) (4.87-11.55%) and Nitrogen (N) contents were also influenced by the industrial wastes and effluents of the factories.

The water soluble fraction is certainly the most biologically active. The high toxicity potential of this fraction is proven by the higher sensitivity exhibited by plants grown in hydroponic media. The water soluble fraction has highest potential of contamination of food chain, surface water and groundwater (Leita and De Nobili, 1991; He *et al.,* 1992).

1st sampling during wet season

The Pb concentration ranges in different extraction fractions of the 1st collected samples, were found to be 1.84-43.35, 2.85-55.0, 6.1-109.91 and 11.0-213.0 mg kg^{-1} Pb in water-soluble, NH_4OAc extractable, DTPA extractable and total fractions respectively (Table II).

The Pb concentration decreased from location 1 to successive locations in all respect but total Pb concentration at sampling points 1 to 12 were found variable and in some cases above the tolerable limit value of 100 mg kg^{-1} (Kloke, 1980).

Water soluble Pb concentrations in different sampling points were significantly different from each. The lowest concentration of water soluble lead was found in the sampling point 14 and 18. On the other hand the highest concentration of water soluble Pb was found in the sampling point 1 i.e. at the source point.

NH_4OAc and DTPA extractable Pb concentration at different sampling points were significantly different from each other. The lowest concentration of NH_4OAc extractable Pb was found at the sampling point 14 and the highest was found in the sampling point 1 i.e. at the source point. The lowest concentration of DTPA extractable Pb was found in the sampling point 14 and the highest was found in the sampling point 1 i.e. at the source point.

In case of total Pb concentration in soil, the lowest value was at location 14 and the highest was in the sampling point 1 i.e. at the source point, which was above the tolerable limit value (Kloke, 1980). Total lead concentration in 24 sampling points differed significantly from each other with some exceptions (Table II).

2nd sampling during dry season

In the 2nd collected samples during dry season, the Pb concentration ranges in different fractions of extraction were found to be 3.21-27.48, 7.30-47.18, 16.26-101.61 and 59.83-222.2 mg kg-1 Pb in water-soluble, NH4OAc extractable, DTPA extractable and total fractions respectively (Table III). The average Pb concentration in different soil fractions were 7.80, 21.85, 56.15 and 130.29 mg kg-1 Pb in water-soluble, NH_4OAc extractable, DTPA extractable and total fractions respectively (Table III). Comparing wet and dry season sampling data, mean Pb concentrations in different soil fractions were higher in the 2nd sampling data with the exception of

Table II: Concentration of lead in soils with different extractions during rainy season, 1st sampling (in mg kg^{-1})

Location	Longitude, North	Latitude, East	Water soluble	NH4OAc Extractable	DTPA	Total Extractable
1	23°45.7	90°24.5	43.35 w	55.0 n	109.91 r	213.00 u
2	23°45.8	90°24.6	11.58 o	25.76 jk	45.00 k	138.00 q
3	23°46.0	90°24.7	21.79 u	41.40 m	76.00 q	154.00 t
4	23°46.2	90°24.8	12.79 q	22.66 ij	38.00 i	98.00 k
5	23°46.3	90°24.9	14.35 r	32.19 l	49.00 l	103.50 m
6	23°46.2	90°25.0	17.06 s	33.80 l	54.00 m	130.00 o
7	23°46.1	90°25.1	17.92 t	24.67 jk	55.00 n	123.00 n
8	23°46.1	90°25.4	24.95 v	42.74 m	70.00 p	143.00 r
9	23°46.1	90°24.9	10.06 m	19.04 li	35.00 h	76.00 i
10	23°45.9	90°24.8	7.57 j	16.57 gh	30.00 g	135.00 p
11	23°45.7	90°24.4	6.55 i	9.39 bcd	28.00 f	143.50 s
12	23°45.7	90°27.8	9.66 l	27.10 k	45.00 k	100.00 l
13	23°45.7	90°27.9	11.00 n	14.00 efg	28.00 f	56.00 e
14	23°45.7	90°28.1	1.84 a	2.87 a	6.10 a	11.00 a
15	23°45.7	90°28.4	12.77 p	24.63 jk	43.00 j	87.00 j
16	23°45.6	90°28.7	11.00 n	12.00 def	24.00 e	73.00 h
17	23°45.6	90°28.7	3.58 d	8.53 bcd	14.00 d	60.00 f
18	23°46.6	90°28.9	2.63 b	6.47 abc	12.00 c	45.00 d
19	23°45.5	90°28.9	3.36 c	5.67 ab	11.00 b	31.00 c
20	23°45.9	90°27.8	3.99 e	5.72 ab	12.00 c	29.00 b
21	23°45.6	90°27.5	5.87 f	15.42 fgh	24.00 e	76.00 i
22	23°45.6	90°27.2	6.30 h	16.50 gh	45.00 k	72.00 g
23	23°46.6	90°26.8	6.23 g	10.42 cde	55.00 n	87.00 j
24	23°45.7	90°26.5	7.96 k	8.30 bcd	59.00 o	98.00 k
Max			43.35	55.00	109.91	213.00
Min			1.84	2.87	6.10	11.00
Mean			11.42	20.27	40.33	95.08

Mean values with the same letter (s) in a column is not significantly different at 5% level by Duncan's Multiple Range Test (DMRT).

water soluble fraction which might be due to flooding or dilution effect during wet season.

The Pb concentration decreased from point source (location 1) to successive locations in all respect but total Pb concentration in most of the cases were found to be above the tolerable limit value (Kloke, 1980).

Water soluble lead concentrations in different sampling points were significantly different from each other except some sampling points. The lowest concentration of water soluble Pb was found in the sampling point 14. On the other hand the highest concentration of water soluble Pb was found at the source point 1.

NH_4OAc and DTPA extractable Pb concentration at different sampling points were significantly different from each other. The lowest concentration of NH_4OAc extractable and DTPA extractable Pb was found at the sampling point 14 and the highest was found in the sampling point 1 i.e. at the source point.

In case of total Pb concentration in soil, the lowest value was in point 14 and the highest was in the sampling point 1 which was above the tolerable limit value (Kloke, 1980). Total lead concentration in 24 sampling points differed significantly from each other except some points (Table III). Total Pb concentrations at some sampling points were above

Table III: Concentration of Pb in soils with different extractant during 2nd sampling

Location	Longitude, North	Latitude East	Water soluble Pb	NH_4Oc Exchbngeble PB	DTPA Extractable Pb	Total Pb by Aqua-regia
			mg kg^{-1}			
1	23°45.7	90°24.5	27.48 c	47.17 c	101.6 c	222.2 e
2	23°45.8	90°24.6	16.73 abc	31.41 bc	62.04 bc	145.86 cd
3	23°46.0	90°24.7	21.15 bc	29.45 abc	70.40 bc	150.48 cd
4	23°46.2	90°24.8	9.18 ab	18.02 ab	45.32 ab	166.54 de
5	23°46.3	90°24.9	11.76 ab	19.14 ab	41.80 ab	94.04 abc
6	23°46.2	90°25.0	3.23 a	7.31 a	16.28 a	59.84 a
7	23°46.1	90°25.1	5.43 a	12.74 ab	35.64 ab	77.88 ab
8	23°46.1	90°25.4	7.59 ab	9.55 ab	76.12 bc	124.54 bcd
9	23°46.1	90°24.9	27.46 c	47.17 c	101.59 bc	222.19 e
10	23°45.9	90°24.8	16.71 abc	31.41 bc	62.03 bc	145.85 cd
11	23°45.7	90°24.4	21.14 bc	29.44 abc	70.39 bc	150.47 cd
12	23°45.7	90°27.8	9.14 ab	18.07 ab	45.31 ab	166.53 de
13	23°45.7	90°27.9	11.75 ab	19.13 ab	41.79 ab	95.03 abc
14	23°45.7	90°28.1	3.21 a	7.30 a	16.26 a	59.83 a
15	23°45.7	90°28.4	5.41 a	12.73 ab	35.63 ab	77.87 ab
16	23°45.6	90°28.7	7.58 ab	9.54 ab	76.11 bc	124.51 bcd
17	23°45.6	90°28.7	27.39 c	47.18 c	101.61 c	222.21 e
18	23°46.6	90°28.9	16.73 abc	31.42 bc	62.05 bc	145.87 cd
19	23°45.5	90°28.9	21.16 bc	29.46 abc	70.41 bc	150.49 cd
20	23°45.9	90°27.8	9.19 ab	18.03 ab	45.33 ab	166.54 de
21	23°45.6	90°27.5	11.77 ab	19.15 ab	41.41 ab	95.05 abc
22	23°45.6	90°27.2	3.23 a	7.32 a	16.29 a	59.85 a
23	23°46.6	90°26.8	5.43 a	12.75 ab	30.65 ab	77.89 ab
24	23°45.7	90°26.5	7.60 ab	9.56 ab	76.133 bc	124.53 bcd
Max			27.48	47.18	101.61	222.21
Min			3.21	7.30	16.26	59.83
Mean			7.80	21.85	56.15	130.29

Mean values followed by the same letter (s) within the column is not significantly different at the 5% level by DMRT.

the maximum tolerable limit (100 mg kg^{-1}) according to Kloke (1980).

As a whole, the Tejgaon industrial area soils, had the highest concentration of total, water soluble, NH_4OAc extractable and DTPA extractable Pb in some points which might be due to discharge of liquid wastes, flocculated sludge and other solids with excessive metals coming from different industrial processes. The highest concentration of Pb and other metals in this area is not only a problem with respect to plant nutrition and the food chain, but also a direct health hazard because of its carcinogenic characteristics. The area is not only occupied by industries but also inhabited by many slums people around the complexes and on the bank of waste discharging channel. Recently many urban area/housing projects namely, Aftab Nagar, Bansree housing project, Niketon project etc. have developed on the bank of waste discharging channel. So, these may be ecological implications associated with metal poisoning to the slum living children because they have direct contact with the contaminated soil and effluents (Thornton *et al.,* 1985). The highest concentration of total and extractable Pb, is also due to effluents and solid wastes of textile dying industry.

Water soluble, NH_4OAc extractable and DTPA extractable Pb have been considered as available Pb (Lindsay and

Norvell, 1978). The sum of these Pb species is a measure of available concentration of Pb in the contaminated soils of Tejgaon industrial area. Considering the critical limits of DTPA-extractant for Pb (Lindsay and Norvell, 1978), some of the sampling points were above the critical limits. The concentration of Pb in the soil solution phase strongly increased with decreasing pH and increasing total content of this metal in the different sampling points. The similar results were also reported by many authors (Chamon *et al.,* 2005, Nuruzzaman *et al.,* 1995, Mondol *et al.*, 2002).

Water samples

From the 1st sampling water data (wet season) TSS and TDS values were much above the maximum standards of water quality (data not given here) (CPCB, 1995; DOE, 1992). BOD, COD and DO values were much lower than the minimum standards of water quality (data not given here) (CPCB, 1995; DOE, 1992). The general water quality of the whole study area was extremely poor. Foul smell was so intense that some of the workers had to abandon the sampling protocol.

Table IV : Concentration of Pb in water samples brought during 1st and 2nd sampling

Sampling Location	Longitude, North	Latitude East	1st sampling	2nd sampling
			mg kg^{-1}	
1	23°45.7	90°24.5	1.469 a	nd
2	23°45.8	90°24.6	1.423 b	nd
3	23°46.0	90°24.7	1.374 c	nd
4	23°46.2	90°24.8	1.285 d	nd
5	23°46.3	90°24.9	1.219 e	nd
6	23°46.2	90°25.0	0.845 f	nd
7	23°46.1	90°25.4	0.85 g	nd
8	23°46.1	90°24.9	0.802 h	nd
9	23°45.9	90°24.8	0.753 i	nd
10	23°45.6	90°24.6	0.681 j	nd
11	23°45.7	90°24.4	0.595 k	nd
12	23°45.7	90°27.8	0.509 l	nd
13	23°45.7	90°27.8	0.452 n	nd
14	23°45.7	90°27.9	0.402 m	nd
15	23°45.7	90°28.1	0.336 o	nd
16	23°45.7	90°28.4	0.269 p	nd
17	23°45.6	90°28.7	0.232 q	nd
18	23°45.6	90°28.7	0.153 r	nd
19	23°46.6	90°28.9	0.06 s	nd
20	23°45.5	90°28.9	0.04 s	nd
21	23°45.55	90°29.0	0.133 t	nd
22	23°45.9	90°27.8	0.212 u	nd
23	23°45.6	90°27.5	0.249 v	nd
24	23°45.6	90°27.2	0.223 w	nd
25	23°46.6	90°26.8	0.316 x	nd
26	23°45.7	90°26.5	0.382 y	nd
27	23°45.8	90°26.2	0.432 z	nd
28	23°45.8	90°25.9	0.489 a	nd
29	23°46.0	90°25.6	0.575 b	nd
mean			0.661	

nd=not done

Mean values with the same letter (s) in a column is not significantly different at 5% level by DMRT.

The concentration of Pb in water sample during rainy season ranges from 0.04 to 1.469 mg L^{-1} (Table IV). The average concentration was 0.661 which was much above the tolerable drinking water standard according to WHO (2004) (Table V). Ullah *et al.* (1995) investigated heavy metal pollution of soils and water and their transfer into plants in Bangladesh. According to Ullah *et al*. (1996) and Chamon *et al.* (2005), Tejgaon industrial area was polluted with Ni, Pb and Cd.

Plant samples

Heavy metal concentration in plant samples was high at the study area. Plant samples accumulated more and tolerated higher amounts of heavy metals as reported by Chamon *et al.* (2005).

Average total concentration of Pb were 19.65 and 19.83 mg kg^{-1} respectively (Table V), during wet and dry season, at Tejgaon industrial area which were in the range of plants general toxic concentration level (Sauerbeck, 1982).

Table V: Concentration of Pb in plant samples brought during wet and dry season

Sampling Location	Plant samples				
	Longitude, North	Latitude East	Sample type	Wet season	Dry season
				mg kg^{-1}	
1	23°45.7	90°24.5	Grass	19.53 abc	20.58 b
2	23°45.8	90°24.6	Bean leaf	14.60 a	20.87 ab
3	23°46.0	90°24.7	Grass	17.60 ab	22.57 b
4	23°46.2	90°24.8	Grass	19.13 abc	17.5 a
5	23°46.3	90°24.9	Grass	17.20 a	20.23 ab
6	23°46.2	90°25.0	Grass	18.67 abc	20.83 ab
7	23°46.1	90°25.1	Water hyacinth	20.53 abc	17.57 a
8	23°46.1	90°25.4	Water hyacinth	24.33 bc	19.53 ab
9	23°46.1	90°24.9	Bean leaf	25.27 c	21.63 b
10	23°45.9	90°24.8	Kalmi	19.54 abc	17.7 a
11	23°45.7	90°27.8	Water hyacinth	14.61 a	20.83 ab
12	23°45.7	90°27.9	Water hyacinth	17.61 ab	20.58 b
13	23°45.7	90°28.1	Bean leaf	19.14 abc	17.6 a
14	23°45.7	90°28.4	Kalmi	17.21 a	20.3 ab
15	23°45.6	90°28.7	Kalmi	18.68 abc	20.9 ab
16	23°45.6	90°28.7	Water hyacinth	20.54 abc	17.6 a
17	23°46.6	90°28.9	Kalmi	24.34 bc	19.6 ab
18	23°45.5	90°28.9	Kalmi	25.28 c	21.7 b
19	23°45.55	90°29.0	Kalmi	19.52 abc	17.6 a
20	23°45.9	90°27.8	Water hyacinth	14.59 a	20.77 ab
21	23°45.6	90°27.5	Water hyacinth	17.59 ab	22.44 b
22	23°45.6	90°27.2	Water hyacinth	19.12 abc	17.4 a
23	23°46.6	90°26.8	Water hyacinth	17.19 a	20.13 ab
24	23°45.7	90°26.5	Kalmi	18.66 abc	20.73 ab
25	23°45.8	90°26.2	Water hyacinth	20.52 abc	17.47 a
26	23°45.8	90°25.9	Water hyacinth	24.32 bc	19.43 ab
27	23°46.0	90°25.6	Water hyacinth	25.26 c	21.53 b
Max				25.28	22.57
Min				14.59	17.4
Mean				19,65	19.83

Mean values with the same letter (s) in a column is not significantly different at 5% level by DMRT

Metal contaminations of soils, sediments, water bodies and plants have also been reported earlier (Bharma and Costa, 1992; Ullah *et al.* 1995, 1999; Chamon *et al.*, 2005 and Kashem and Singh, 1998). Concentrations of total soil Pb in most of the locations were found to be above the toxic level (Table II and III). During 2nd sampling Pb concentration in around 50% plant samples were higher than 1st sampling data (Table V). Pb content was above the critical limit in grass and water hyacinth in some sampling points e.g in the point 7, 8, 9, 16, 17, 18, 25, 26 & 27 but low in other plant samples. During the 2nd sampling Pb concentrations in all plant samples, except a few, were above critical limit.

Extent of contamination by Pb in soils, plants and water:

Expeditions to collect samples were extremely difficult due to the foul smell and unknown nature of gases emitting out of the water body. The soil, water and air were unfit for any kind of human consumption. But a vast number of people are living and working in the area disregarding the existing severe health hazards. Results show that the soil, water and plant were of extremely low quality in the rainy season. Their quality deteriorated several fold in the dry season.

The pH dependence of the soluble Pb content show that the mobility and availability of the metal is high at strongly to extremely acid soil reaction. This was also reported by Brümmer *et al.,* (1986).

The extent of Pb contamination due to heavy metal deposition in Tejgaon industrial sites are presented in Table VII. The total Pb content of the soils, water and plant samples showed wide range of values from background to a level considered to reflect severe contamination. The extent of

Table VI: Extent of contamination according to total Pb concentration in soils, water and plants (mg kg^{-1}) during wet and dry season sampling. Percentage of sampling locations in parentheses

Metals		Group 1	Group 2	Group 3
Soil (n=24)				
	Water soluble	≤20 (87.5)	>20-≤100 (12.5)	≥100(0)
Pb (wet season)	NH_4OAc	≤20 (58)	>20-≤100 (42)	≥100 (0)
	DTPA	≤20 (21)	>20-≤100 (75)	≥100 (4)
	Total	≤20 (4)	>20-≤100 (58)	≥100 (38)
	Water soluble	≤20 (75)	>20-≤100 (25)	≥100 ((0)
Pb (dry season)	NH_4OAc	≤20 (62)	>20-≤100 (38)	≥100 (0)
	DTPA	≤20 (12.5)	>20-≤100 (75)	≥100 (12.5)
	Total	≤20 (0)	>20-≤100 (37.5)	≥100 (62.5)
Water (n=29)				
	Pb (wet season)	0.00-< 0.01 (0)	0.01(0)	>0.01 (100)
	Pb (dry season)	nd	nd	nd
Plant (n=27)				
	Pb (wet season)	0-<10 (0)	>10- ≤20(67)	>20 (33)
	Pb (dry season)	0-<10 (0)	>10- ≤20(41)	>20 (59)

Group 1 = Background level, group 2= Maximum tolerable level and group 3 = In excess of tolerable level.

Soil

Background concentration for Pb = 12-20 mg kg^{-1} (Singh and Steinnes, 1994) of Bangladesh soil. Tolerable total concentrations is 100 mg kg^{-1} for Pb (Kloke, 1980).

Water

Background concentration for Pb =0.00-< 0.01 mg L^{-1} ppm of drinking water of Bangladesh, (WHO, 2004). Tolerable total concentrations is >0.01 ppm for Pb (WHO, 2004).

Plant

Background concentration for Pb = 0-<10 mg kg^{-1} (Sauerbeck, 1982). Toxic concentrations is10-20 mg kg^{-1} for Pb (Sauerbeck, 1982).

contamination (in %) was identified by using information of background levels of total Pb in soils (Singh and Steinnes, 1994) and background levels of Pb for Bangladesh soils (Domingo and Kyrzma,1983). The tolerable and in excess of tolerable level was calculated on the information of Kloke (1980).

Background concentration for Pb of Bangladeshi soils is 20 mg kg^{-1} (Domingo and Kyuma, 1983). Tolerable total concentration of Pb in soil is 100 mg kg-1 (Kloke, 1980). The total Pb concentration of the soil samples at different points showed wide range of values from background to a level considered to reflect severe contamination. The extent of contamination was identified by using information of background levels of total Pb in soils (Domingo and Kyruma, 1983; Singh and Steinnes, 1994; Nuruzzaman et al. 1995; Ullah *et al.* 1999 and Chamon *et al.* 2005). The tolerable and in excess of tolerable level was calculated on the information of Kloke (1980).

Concentrations of total Pb in soils during 1st sampling were found above the natural background level with few exceptions. Only 4% samples contained total Pb in the normal range (=20 mg kg^{-1}). Total Pb concentration in the group of tolerable level was 58% (>0.220=100 mg kg^{-1}) and 38% (=100 mg kg^{-1}) soil samples were found in the group of in excess of tolerable level. In case of dry season no soil sample was found to contain Pb in the normal range (=20 mg kg^{-1}). Total Pb concentration in the group of tolerable level(>0.220=100 mg kg^{-1}) was 37% and 63% soil samples were found in the group of in excess of tolerable level (=100 mg kg^{-1}). Different solvent extractable Pb concentrations were also found variable at successive sampling locations as shown in table II and III.

In case of water samples, Pb concentration in the rainy season, all samples were found in the group of in excess of tolerable level (>0.01 mg L^{-1}) (Table VI).

During 1st sampling period, rainy season saw dilution of water due to influx of rain water and floodwater from surrounding areas. Fresh water diluted and washed away much of the pollutants. But it was not enough to completely wash pollutants away. As water receded and pollution load increased with dry season (2nd sampling period), heavy metals concentration increased 2 to 3 times compared to rainy season (Ullah *et al.,* 1999).

Plants take up heavy metals from solution without any hindrance. During the rainy season most plants were grass and water hyacinth with a few other plants. Most of the grasses and water hyacinth find their way into the food chain in the form of cattle feed. During the dry season rice, grass and many other types of 'rabi' crops, e.g., tomato, capsicum, cabbage, cauliflower, etc. were growing in the contaminated soils irrigated with these polluted water. Pb concentrations were very high and above critical level in all crop plants in both seasons. 33 and 59% of plant samples were found in the group of in excess of tolerable level during wet and dry season, respectively (Table VII).

One of the most alarming facts is that some sampling points under investigation were apparently more polluted than sampling point near pollution source. Bumper crops were being produced in the vicinity of Balu river. Locally produced crops with attractive appearances may have high Pb content.

Concentrations of heavy metals such as Pb was much higher in grass and water hyacinth in the Tejgaon industrial area and it may be due to the exposure of these plants to heavy metals for a longer period or there might be different mechanism of absorption of heavy metals by these plants. Elevated levels of Cd, Cu, Pb and Zn in plants in this area have been demonstrated by other investigators (Kashem and Singh, 1998; Ullah *et al.,* 1999; Mondol *et al.,* 2002; Chamon *et al.*, 2005).

Distribution of total (HNO_3-HCl) Pb with distance

Distribution of water soluble, NH_4OAc extractable, DTPA extractable and total lead content in the soils of Tejgaon Industrial area during wet and dry season are presented in Fig. 1 and 2. The Pb concentrations in soils with different extractant were found to decrease with distance away from the disposal point due to dilution of the effluent and water. Similar findings were also observed by other investigators. Freedman and Hutchinson (1980) and Blom (1986) reported a very drastic decrease in metal concentration away from the source point i.e from an electro-metallic industry in southeastern Norway. The decreasing tendency with distance indicates the accumulation of metals by these industrial operations. The Tejgaon industrial area is low-lying and wet through most part of the year. This part is influenced by the battery, biscuit, paper and pulps, textiles, carbides, pharmaceuticals, pesticides, distilleries etc, where all effluents and wastes converge and the zone under investigation is designated as the confluence zone. Waste effluents discharged by these industries heavy metal concentration in environmental samples in this area. Low pH values in the respective sites could possibly increase solubility and mobility of metals in the soils. Increased solubility of metals is likely to lead to

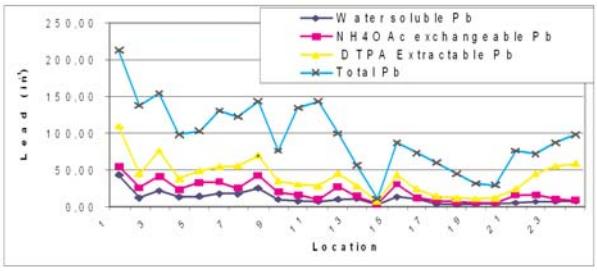

Fig. 1: Distribution of Pb in different fractions with distance (not to the scale) during wet season

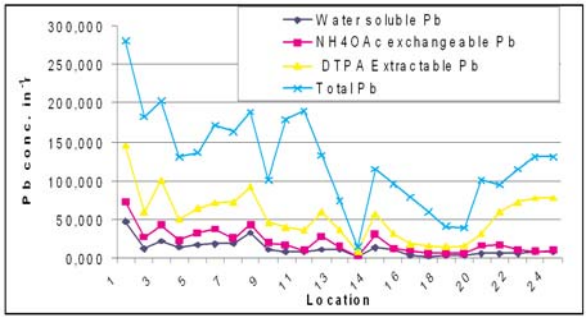

Fig. 2: Distribution of Pb in different fractions with distance (not to the scale) during dry season

increased availability of the metals for plant uptake, and it might cause Pb toxicity. This results indicate that appreciable soil acidification has taken place in the vicinity of that area. This is in agreement with the result from other investigators (Kashem and Singh, 1998; Mondol *et. al.,* 2002; Chamon *et al.,* 2005).

Interrelationships among the water soluble, NH_4OAc extractable, DTPA extractable and total Pb in the Tejgaon industrial soils during wet and dry season

A correlation matrix for Pb metal in the soils of Tejgaon industrial sites was calculated to see if some of the Pb fractions were interrelated with each other and the results are presented in Table VIII and IX.

A number of fractions were observed to be significantly ($p<0.01$) correlated with each other. For example, water soluble fraction of Pb species was positively and significantly correlated with all other species of Pb. The total contents of Pb were found to be highly correlated with their extractable amounts.

Soil NH_4OAc and DTPA extractable species of Pb were found to be significantly correlated with other species of Pb during both wet and dry season (Table VII and VIII). Correlation between Pb content in soil and plant is insignificant.

Table VII: Descriptive statistics and Correlation matrix for different lead extractable species in soil and their correlation with plant Pb during wet season (n=24)

Descriptive Statistics

	Mean	Std. Deviation
Water soluble	11.4233	9.05762
NH_4OAc	20.0354	13.47697
DTPA	40.3338	24.15164
Total	95.0833	46.74228
Plant	19.1867	3.05353

Correlations

	Water soluble	NH_4OAc	DTPA	Total	Plant
Water-soluble	1	922(**)	886(**)	.808(**)	.087
NH_4OAc		1	.865(**)	.816(**)	.049
DTPA			1	.845(**)	-.029
Total				1	-.106
Plant					1

** Correlation is significant at the 0.01 level (2-tailed).

Table VIII: Descriptive statistics and Correlation matrix for different lead extractable species in soil and their correlation with plant Pb during dry season (n=24)

Descriptive Statistics

	Mean	Std. Deviation
Water soluble	12.8117	7.97006
NH_4OAc	21.8521	12.80986
DTPA	55.9413	25.82297
Total	130.2529	50.33067
Plant	19.8804	1.67106

Correlations

	Water soluble	NH_4OAc	DTPA	Total	Plant
Water-soluble	1	.970(**)	.826(**)	.837(**)	.245
NH_4OAc		1	.762(**)	.842(**)	.243
DTPA			1	.856(**)	.150
Total				1	.169
Plant					1

** Correlation is significant at the 0.01 level (2-tailed).

Correlation between lead metal species in soil and in Plants

Mean, standard deviation and correlations (r) in plants Pb with different soil Pb species are presented in table 7 and 8 during wet and dry season.

During wet season plant Pb positively correlate only with water soluble and NH_4OAc extractable Pb species and negatively correlate with DTPA and total species of Pb in soil.

During dry season plant Pb positively correlate with all species of Pb in soil i.e. plant Pb positively correlate with water soluble, NH_4OAc extractable, DTPA extractable and total Pb species in soil (Table VIII).

The positive relationship among the soil and plant cotent metal might be a cause of Pb toxicities to both plants and animals through their entry into food chain (Sameni *et al.,* 1987; Roads *et al.,* 1989).

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