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Full Length Research Paper

An evaluation of soil contamination with metals and hazardous industrial waste disposal site located in Hyderabad city

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Geo-environmental evaluation of heavy metals in/and around hazardous waste disposal sites located in the north-western part of Hyderabad (India) was carried out to define the degree of contamination of soil environment. Approximately 50,000 tonnes per annum of hazardous/industrial waste is deserted as landfill over 200 acres of area in the city outskirts, contaminating soil resource. In the present study, heavy metals (As, Cr, Cu, Ni, Pb, Zn) in soil samples were analysed by X-ray fluorescence spectrometer to quantify their concentrations. Natural background values were used to delineate their derivation as geogenic or anthropogenic. The average concentrations of As, Cr, Pb was found to exceed the threshold and natural background values, whereas the upmost concentrations of Cu, Ni and Zn exceeded the prescribed threshold limit. Soil pH varies from 5.7 to 8.9 and is acidic to near neutral and alkaline in nature. Soil pH significantly affects the solubility and mobility of these metals, as most of the metals are soluble in acid soils than in neutral or slightly basic soils. The methodology used has proved to be a useful tool to separate geological and anthropogenic causes of variation in soil heavy metal content and to identify common pollution sources.

Keywords: Industrial waste, landfill, heavy metal, geo-environment, soil, Hyderabad.

INTRODUCTION

Physicochemical properties of soils depend on both natural and anthropogenic factors, together acting over different spatial and temporal scales. Natural pedological processes (rock weathering and organic matter decomposition) are related to parent material, geomorphology of the area, presence of vegetation, the climatic conditions and other interactions with the environment. The effects of these processes are strictly time-dependent and exposed in a quite complex structure of soils. In contrast, soil management practices significantly affect pedological properties by changing soil structure mechanically due to agricultural and urban activities, and by changing chemical composition through pollution load. The presence of any element in a fatal concentration in the soil could be due to both natural and anthropogenic factors; therefore it is often quite difficult to discriminate among the different causes. The parent material largely influences heavy metal content in many soil types, with concentration sometimes exceeding the critical values (Palumbo et al., 2000; Salonen and Korkka-Niemi, 2007). Several heavy metals, such as Ni, Cr and Mn, are contained as trace elements in some rock types of volcanic and metamorphic origin (Alloway, 1995a). During weathering processes the primary crystalline structures of some rock minerals are completely broken and relevant chemical elements are thus either adsorbed in the topsoil or transported towards surface water or groundwater targets. Anthropogenic sources of heavy metal in soils are mainly hazardous/solid waste disposal and combustion processes in industry and transportation. Moreover, mining activities for extraction and manufacturing of metal products may result in a large amount of pollutants being released into the atmosphere and, secondly, in the adjoining soils and waters. Long-term and extensive use

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Table 1: Content of Various Elements in Soils

Metals	Selected Average for Soils mg/kg	Common Range for Soils mg/kg	Natural Background Values of Metals in Granitic Soil of Hyderabad City mg/kg
Al	71,000	10,000-300,000	-----
Fe	38,000	7,000-550,000	-----
Mn	600	20-3,000	-----
Cu	30	2-100	14 to 33
Cr	100	1-1000	35 to 93
Cd	0.06	0.01-0.70	0 to 0.2
Zn	50	10-300	34 to 64
As	5	1.0-50	13 to 36
Se	0.3	0.1-2	-----
Ni	40	5-500	19 to 57
Ag	0.05	0.01-5	-----
Pb	10	2-200	35 to 94
Hg	0.03	0.01-0.3	-----

Source: (Lindsay, 1979; Murthy, 2008)

of agricultural land with frequent application of pesticides (Nicholson et al., 2003) will result in heavy metals such as copper, nickel, zinc and cadmium accumulating in the topsoil. A soil pollution assessment becomes very complex when different sources of contamination are present and their products are variably distributed. In these cases the spatial variability of heavy metal concentrations in soils is basic information for identifying the possible sources of contamination and to delineate the strategies of site remediation. A detailed appraisal of the characteristics of urban soils points out that soils in urban and suburban areas are frequently disturbed and subjected to mixing, filling and contamination with inorganic components and organic residue (Craul, 1985). Hazardous waste disposal sites are one of the major sources of elevated levels of metals in the soil environment. Migration of contaminants from waste disposal sites to surrounding ecosystems is a complex process and involves various geochemical activities. Soil pH considerably affects the solubility of metals. Besides pH, the solubility of metals is strongly influenced by the redox potential, the presence of complexing agents such as chlorides, sulphates, carbonates and organic acids, and the properties of the solid waste phases in or on which metals can be bound (Bozkurt et al., 2002). These metals can bio-magnify in plants and animals eventually making their way to humans through the food chain (Abrahams, 2002). Most of the metals are soluble in acid soils than in near neutral, neutral or slightly basic soils (Schmitt and Sticker, 1991). Iron, manganese and lead have low to very low mobility at pH < 7 and thus, would be enriched in soil. Nickel, copper and zinc have high mobility under acidic conditions and due to formation of sparingly soluble metal sulphides with very low mobility under reducing conditions, these metals in soils can either be enriched or depleted relative to parent material depending on the dominant factors that exist in the weathering environment (Mattigod and Page, 1983). Depending upon the local geology, the concentration of metals in soil may exceed the common ranges (Lindsay, 1979), (Table 1). Use of common ranges or average concentrations of trace metals in soils as an indicator of whether a soil is contaminated, is not appropriate since the native concentration of metals in a specific soil may fall out of the listed ranges. Only by direct analysis of uncontaminated soils can background levels of metals be determined. The natural background value (BGV) of metals in soil of Hyderabad city was established by analysing soils from the uncontaminated regions (Murthy, 2008), (Table 1) and used as a sign of whether the soil around study area is polluted. The present study was taken up to establish the levels of potentially toxic elements in soil environment around hazardous waste disposal site in Hyderabad city. In this case, the BGV is used as a tool to differentiate the source of metals in soil as anthropogenic or geogenic.

MATERIALS AND METHODS

Study Area

The dumpsite is located in the north-western part of Hyderabad (Figure 1) and lies between the Longitudes 78°59′ N and Latitude 17°51′ E. It extends over 200 acres of area and receives on an average 200-300 tons of industrial solid waste per day (~50,000tons/annum). It is a part of peninsular gneissic complex, and the area around the dumpsite is covered by granite, which is a fraction of large granitic batholiths having exposures covering an area over 5000 km^2 belonging to the Archaean age (Janardhan, 1965; Kanungo et al., 1975; Pandey et al., 2002). The granites are medium to coarse grained and are mainly of two types: grey granite and pink granite – depending upon the colour of the feldspars. Mineralogically, these rocks consists of quartz (21%-42%), potash

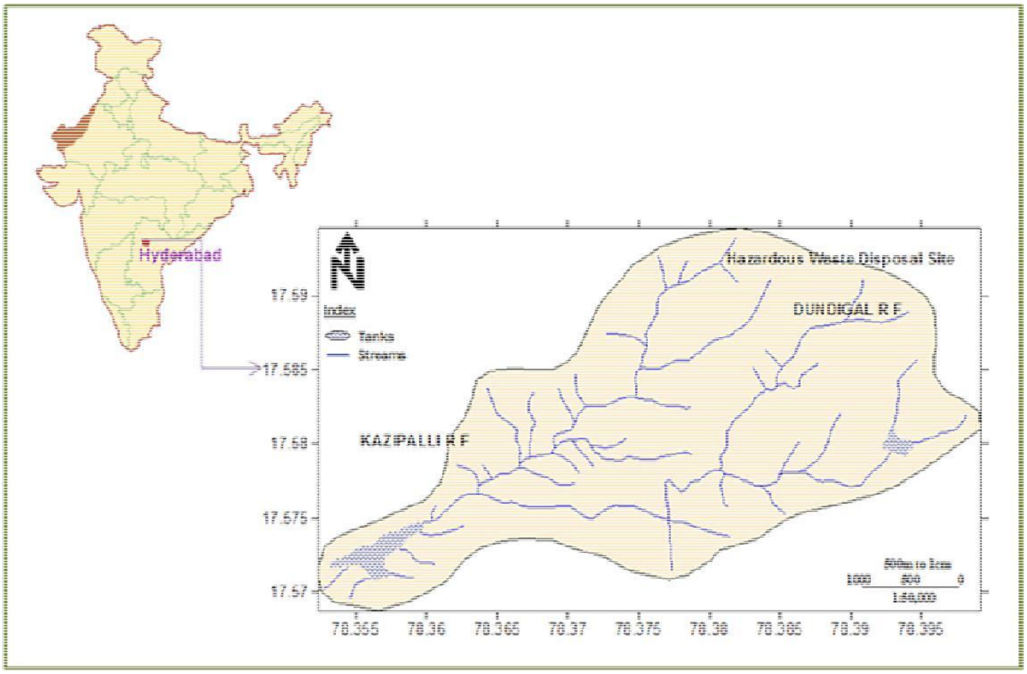

Figure 1: Key map of the study area

feldspar (34%-60%) and plagioclase (2%-30) with biotite (1%-22%) forming the chief accessory. Epidote and pyroxene are frequently observed in such terrain (Gnaneshwar and Sitaramayya, 1998). Major part of the study area is covered with pedi-plain having shallow weathering. The soil cover is well-developed residue of weathered granite consists of clay loam, red loam and sandy loam with variable width. The area is semi-arid with subtropical climatic conditions. The temperature varies between 25 °C to 45 °C. It receives more than 80% precipitation from SW monsoon with an average rainfall of 812 mm.

Sample Collection

A systematic soil-sampling program was conducted which includes a total of 45 soil samples. To avoid influence from various arbitrary surface conditions like waste and humus and to assure natural in-place soil, the selected depth of sampling is from 10 to 15 cm below the surface to 25 cm depth. Normally, anthropogenic pollutants contaminate the upper layer of soil. In case of natural pollutant, the entire soil at all depths shows high level of metal enrichment. The samples were taken from geographically distributed (North, South, East and West) sites at target intervals of 300 to 500m in a network formation (Figure 2). Sampling was carried out by using plastic equipment instead of metal tools to avoid any cross contamination. The samples were collected in self-locking polythene bags and were sealed to avoid leakage. The soil samples were air dried and kept in oven for 48 hours at 60 °C. The dried samples were then disaggregated with mortar and pestle and were finely powdered to –250 mesh size (US standard) using a swing grinding mill to make the sample homogeneous. In homogenised sample, the surface layer should represent the bulk specimen; it is an essential step to get accurate analytical data. pH of soil suspension using 1:1 soil to water ratio was determined by the method recommended in Soil Survey Manual, 1993. Weighing of sample was accomplished using analytical balance with precision as low as 0.0001g. Pressed pellets for X-ray fluorescence spectrometer (XRF) analysis were prepared using collapsible aluminium cups (Govil, 1985), with a backing of boric acid. They were then pressed at 25 tons under a hydraulic press to obtain a pellet.

Instrumentation

Elemental composition in soil samples were determined using an X-ray fluorescence spectrometer, type Philips MagiX PRO model PW 2440 XRF with a Rhodium (Rh) anode 4 kW tube. Its high level performance enables a very sensitive and accurate determination of trace and major elements (As, Cr, Cu, Ni, Pb and Zn). With the PW 2440 it is possible to scan the elements of interest from Boron to Uranium. The MagiX PRO is a sequential instrument with a single goniometer-based measuring channel covering the complete elemental array. Suitable software “super Q” was used to take care of dead time correction and inter-element matrix effects. International Soil Reference Materials from the US Geological Survey, Canadian Geological Survey (SO-1, SO-2, SO-3, SO-4), International Working Group France (Govindaraju, 1994), and National Geophysical Research Institute, India (NGRI-D and NGRI-U) (Govil, 1993; Govil and Narayana, 1999), were used to prepare the calibration curves for major and trace elements and to check the accuracy of the analytical data (Govil, 1985; Rao and Govil, 1995).

RESULTS AND DISCUSSION

The individual data acquired for each element are shown in Table 2. The statistical analysis results are summarized as minimum, maximum, average, median and standard deviation of As, Cr, Cu, Ni, Pb and Zn conc-

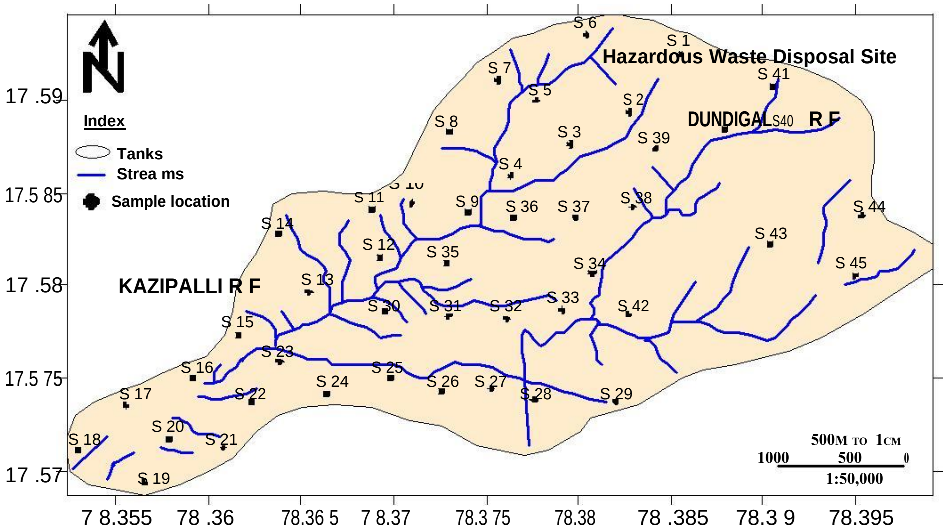

Figure 2: Watershed showing sample location

Table 2: Analytical results of soil-pH and XRF data of heavy metal in soil (mg/kg)

S ID	Soil pH	As	Cr	Cu	Ni	Pb	Zn
S 1	6.5	26.65	86.76	46.46	52.65	74.72	109.88
S 2	7.2	17.65	65.56	20.06	24.75	62.62	102.28
S 3	6.3	276.95	72.96	23.06	25.35	652.32	47.38
S 4	6.2	183.15	211.36	38.06	67.25	436.32	122.08
S 5	5.9	411.45	411.76	44.36	74.55	951.62	112.98
S 6	6.5	40.65	78.56	30.56	50.35	118.92	70.48
S 7	6.1	42.05	68.96	30.26	46.35	124.22	67.38
S 8	7.8	23.35	69.56	40.76	51.25	76.12	77.18
S 9	8.0	77.65	47.66	22.46	26.75	197.02	48.88
S 10	8.8	56.75	102.46	23.46	39.45	154.62	49.88
S 11	7.8	79.55	69.36	16.96	28.75	200.52	78.78
S 12	5.8	59.15	335.76	23.76	74.95	154.22	77.38
S 13	8.1	172.35	159.06	34.36	58.75	420.82	58.28
S 14	7.6	14.45	73.86	22.76	28.85	54.92	62.58
S 15	8.1	17.75	62.66	51.86	41.65	58.22	58.28
S 16	7.9	12.45	72.16	28.86	42.15	42.92	26.98
S 17	8.3	9.95	12.26	11.06	12.55	57.42	97.98
S 18	6.6	18.35	33.46	23.56	25.35	72.22	134.38
S 19	7.7	70.85	426.56	21.96	85.45	169.72	132.28
S 20	8.5	19.25	71.56	41.66	55.05	70.52	36.48
S 21	7.6	19.45	49.46	27.36	29.65	72.12	61.98
S 22	7.4	12.75	56.46	23.26	26.05	50.72	40.78
S 23	7.9	110.95	376.56	53.06	31.15	275.72	54.78
S 24	7.7	15.95	72.16	33.86	48.75	63.72	85.38
S 25	8.4	45.35	112.66	55.76	59.15	118.22	108.58
S 26	6.9	16.55	54.36	24.46	33.05	63.62	86.48
S 27	8.5	13.75	56.36	24.66	30.95	53.82	74.98
S 28	8.2	22.85	88.86	46.56	68.15	70.02	42.38
S 29	7.3	24.25	81.56	40.06	51.95	78.02	75.08
S 30	7.0	24.45	64.46	33.36	39.95	81.02	59.48

Table CONT.

S 31	7.1	26.55	54.96	36.06	44.05	81.12	56.28
S 32	8.8	124.35	309.16	47.36	16.85	308.12	51.08
S 33	8.4	16.25	28.36	16.66	40.75	61.42	49.18
S 34	8.6	13.25	141.16	23.16	24.85	205.42	110.88
S 35	7.9	6.27	403.96	48.16	38.55	488.32	47.08
S 36	6.9	14.45	67.26	20.06	31.75	146.72	345.58
S 37	8.1	7.75	44.56	17.76	131.95	174.72	391.18
S 38	8.9	20.85	106.96	22.66	107.95	160.52	882.18
S 39	6.9	9.65	48.26	186.66	123.75	99.72	312.18
S 40	5.9	18.45	480.56	72.16	72.95	1833.52	279.98
S 41	5.7	6.15	75.56	18.66	27.85	115.02	265.18
S 42	7.5	19.45	149.76	29.86	41.25	223.92	206.88
S 43	7.0	76.15	168.96	31.06	79.65	199.62	95.48
S 44	7.9	19.45	67.46	30.06	20.05	64.32	105.08
S 45	7.4	10.95	63.56	25.46	28.65	52.92	45.98

Table 3: Summary of heavy metal concentrations in soil (mg/kg)

	pH	As	Cr	Cu	Ni	Pb	Zn
Minimum	5.7	6.1	12.2	11.0	12.5	42.9	26.9
Maximum	8.9	411.4	480.5	186.6	131.9	1833.5	882.1
Average	7.5	51.7	127.9	35.2	48.0	206.4	122.3
Median	7.6	19.5	72.2	29.9	41.3	115.0	77.2
Standard Deviation	0.9	77.1	122.4	26.3	26.7	304.3	143.5
Threshold Value*	6 to 8	12.0	64.0	63.0	50.0	70.0	200.0

*Maximum permissible concentrations as defined by CCME 1999

entration (in mg/kg) in Table 3. Maximum permitted threshold concentrations of potentially toxic metals prescribed by CCME, 1999, are also given. The metals concentrations were found to be: Arsenic (6.1-411.4 mg/kg), Chromium (12.2-480.5 mg/kg), Copper (11.0-186.6 mg/kg), Nickel (12.5-131.9 mg/kg), Lead (42.9-1833.5 mg/kg) and Zinc (26.9-882.1 mg/kg). The uppermost concentration of each element fall out of range of natural background values (Table 1), and exceeds the permitted threshold limit. The heavy metals with enrichment levels exceeding the normally expected distribution in soil give rise to concern over the suitability of soil for growing crops (Alloway, 1995b).

The average concentration of Arsenic (As) exceeds the highest background level of 36 mg/kg and threshold value of 12 mg/kg (Tables 1 and 3). Since As exceed the maximum contaminant level (MCL) and natural BGV, the soil is said to be polluted and its source can be ascribed as a consequence of disposal of hazardous/chemical waste (anthropogenic). The maximum concentration of As (411.4 mg/kg) is found in the sample point 5 with soil pH 5.9 (acidic) (Figure 3). Since As is relatively mobile in pH 6.7-8.8 and moderately mobile in pH < 6.6 (Table 4), it is enriched in soils of the study area due to complimentary soil pH condition (5.7-8.9). In the soil environment arsenic exists as either arsenate, As (V) (AsO_4^{3-}), or as arsenite, As (III) (AsO_2^{-}). The toxicity of an arsenic-containing compound depends on its valence state (zero-valent, trivalent, or pentavalent), its form (inorganic or organic), and factors that modify its absorption and elimination. Inorganic arsenic is generally more toxic than organic arsenic, and trivalent arsenite is more toxic than pentavalent and zero-valent arsenic under favourable pH conditions.

Average concentration of Chromium (Cr) exceeded the highest natural BGV of 93 mg/kg and threshold value of 64 mg/kg (Tables 1 and 3). Average soil pH of 7.5 is complimentary for the Cr mobility (Tables 3 and 4), and its high concentration can be seen at regular intervals (Figure 3). It enters the environment through natural and anthropogenic sources. Among natural sources, volcanic emission, bio cycling, weathering of rocks constitute 10 tonnes of Cr per year globally. Anthropogenic input of Cr comes from solid wastes, where approximately 30% of Cr originates from plastic packaging such as plastic shopping bags (Jung et al., 2006). Waste consisting of

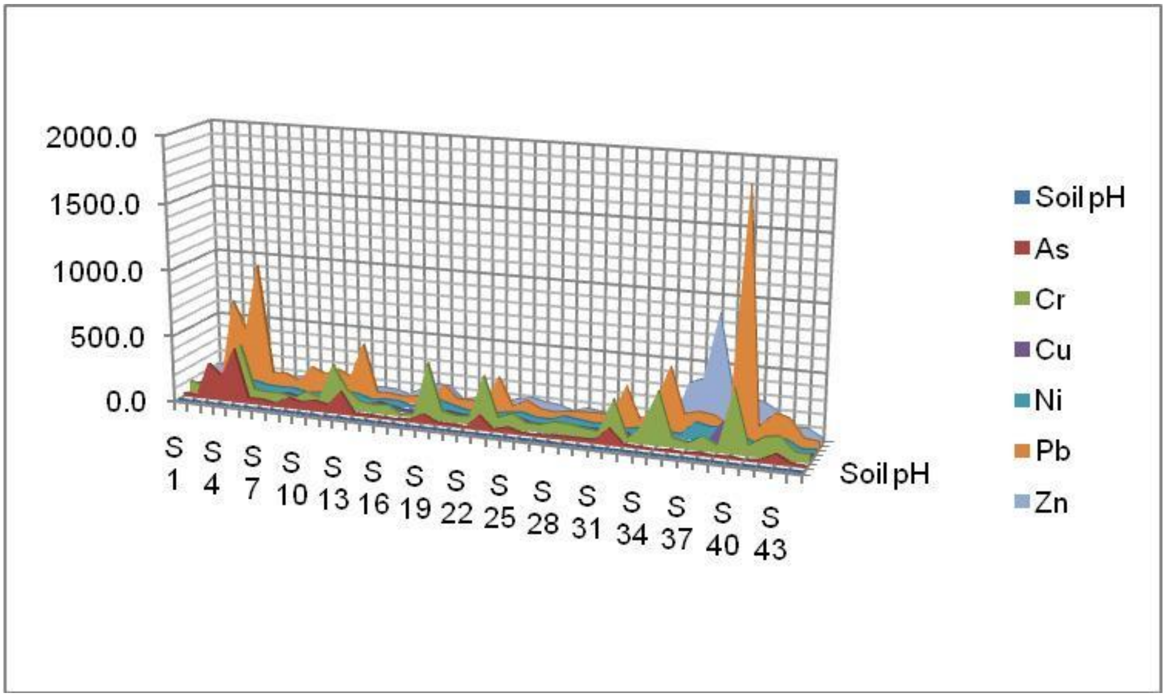

Figure 3: Area chart showing metal concentrations in soil samples

Table 4: Correlation between metal mobility and pH

Mobility	pH 4.2-6.6	pH 6.7-8.8
Relatively mobile	Cd, Hg, Ni and Zn	As and Cr
Moderately Mobile	As, Be and Cr	Be, Cd, Hg and Zn
Slowly / Slightly mobile	Cu, Pb and Se	Cu, Pb and Ni

Source: (Schmitt and Sticker 1991)

lead-chromium batteries, coloured polythene bags, discarded plastic materials and empty paint containers are said to be huge source of Cr. Chromium and its compounds are extensively employed in leather processing and finishing, in the production of refractory steel, electroplating cleaning agents, catalytic manufacture, etc. (Shanker et al., 2005). Because Cr concentration in soil exceeds the highest BGV, its origin can be ascribed to disposal of industrial solid waste over a longer period in the study area.

The highest concentration of Nickel (Ni) (Table 3) exceeds the natural BGV (Table 1), whereas its average concentration is within the threshold value of 50 mg/kg (Table 3). As its concentration level exceeds the BGV and threshold value in some pockets (S4, S5, S12, S13, S19, S20, S25, S28, S37, S38, S39, S40 and S43) downstream of the watershed (Figure 2), its origin can be attributed to industrial solid waste brought to the dumpsite. Depending upon the origin of the soil and pedogenic processes, the surface, or the subsoil may be relatively enriched or have the same Ni concentrations (Adriano, 1986). The +2 oxidation state is the only stable form of nickel in soil environment. Low soil pH favours the exchangeable and soluble Ni^{+2}. Solubility decreases noticeably at higher pH value. Mobility of Ni is medium in acid soils and becomes very low in neutral to alkaline soils (Table 4). Increasing soil pH by adding lime is an effective and practical means of ameliorating the toxicity of Ni^{+2} in soils (Mc Grath, 1995).

Zinc(Zn) ranges from 26.9-882.1 mg/kg. Though its average concentration is within the threshold value (Table 3), the sample points (S36-42) exceeds the threshold and BGV downstream. There is a significant contamination of soil with Zn, exceeding natural BGV throughout the area with highest concentration at sample No. 38 (Figure 3). Zinc in soil solutions exists in +2 oxidation state. Under acidic, oxidizing conditions, Zn^{+2} is one of the most soluble and mobile trace metals. Clay, hydrous oxides and organic matter adsorb Zn strongly, especially under alkaline conditions. The more usual tendency is that, the Zn concentrations are high in surface soils and the concentrations decrease with depth.

Table 5: Correlation coefficients between metals

Elements	As	Cr	Cu	Ni	Pb	Zn
As	1					
Cr	0.42	1				
Cu	0.00	0.16	1			
Ni	0.08	0.23	0.46	1		
Pb	0.47	0.68	0.20	0.19	1	
Zn	-0.14	0.01	0.11	0.56	0.13	1

High potential solubility of Zn^{+2} in acid soils, and the fact that Zn^{+2} is typically a high concentration pollutant of industrial wastes and sewage sludge, combine to create a significant potential for phyto-toxicity from land application of wastes. The chief pollution sources of Zn in soils are metalliferous mining activities, agricultural use of sewage sludge and the use of agro-chemicals such as fertilizers and pesticides. Disposal of chemical waste is a source of abnormal levels of Zn in the study area. Large concentrations of Zn in the soil have adverse effects on crops, livestock and human (Kiekens, 1995).

The average concentration of Copper (Cu) (35.2 mg/kg) lies within the threshold value (Table 3), but exceeds the highest background value of 33 mg/kg (Table 1). Sample points (S39-40) located downstream (Figure 2) show Cu exceeding the threshold concentration. Copper is associated with organic matter, oxides of iron and manganese, silicate clays, and few other minerals. Copper is specifically fixed or adsorbed in soils and is one of the least mobile heavy metals at any pH (Table 4, Figure 3). Organic matter is invariably the dominant factor controlling Cu retention. Most of the total dissolved Cu in surface soils occur in the form of Cu^{2+} organic complex at higher pH condition. It builds up in the surface of contaminated soils showing virtually no downward migration. In alkaline soils Cu^{2+} solubility is very low and the solubility of total copper is enhanced by soluble complexes of Cu^{2+} (mostly hydroxy, carbonate and organic matter complexes). Consequently, mobility may be significant under maximum pH conditions (Mc Bride, 1994). Copper content in the soils largely depends upon the nature of the parent rock, weathering of biotite, orthoclase and plagioclase feldspars and soil characteristics.

Soil Lead (Pb) content varies from 42.9-1833.5 mg/kg. The average concentration of Pb (206.4 mg/kg) exceeds the highest natural BGV (Table 1) and threshold concentration (Table 3). Lead in soil exists in the +2 oxidation state. As soil pH raises, Pb^{+2} ion becomes less soluble under oxidizing conditions. At higher pH, it forms complex with organic matter, chemisorptions on oxides and silicate clays and precipitates as carbonate, hydroxide or phosphate. High Pb content can be seen in maximum number of samples at various pH conditions (Figure 2). Elevated levels of Pb can be attributed to dumping of industrial/chemical waste (anthropogenic).

In order to establish inter-element relationships in soil samples, correlation coefficient for metals were calculated and are shown in Table 5. A very significant correlation was found between Cr-Pb (0.68) and Ni-Zn (0.56), which indicates a common source for these metals. Considerable correlation can be seen between As-Cr (0.42), As-Pb (0.47) and Cu-Ni (0.46), indicating a common origin. Lead is positively correlated to other metals; its load induces the load of others and vice versa. Therefore, Pb levels are directly proportional to the levels of the metal present.

CONCLUSIONS

The present paper aims to evaluate soil contamination with metals in/and around largest hazardous/industrial waste disposal site located in Hyderabad city. Analysis of soil samples from 45 sampling points in the surrounding areas of dumpsite showed significant spatial variation of heavy metals (As, Cr, Cu, Ni, Zn and Pb). The results of the study revealed that soils in the downstream and vicinity of dumpsite are considerably contaminated by metals with their concentrations beyond threshold values. The soil-pH is acidic to alkaline and is one of the major factors affecting mobility/solubility of metals in soil environment. The soils have the potential to retain further additional heavy metal loads in such pH conditions. These heavy metals have a tendency to bio-magnify and induce long-term adverse impact on ecosystem in terms of biochemical and toxicological effect on human being and other composition of our planet. To evaluate the impact of dumpsite and the magnitude of metals leaching out of wastes, this study recommends periodic monitoring of soil environment for toxic metals in the study area.

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