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Heavy Metal Contamination in Roadside Soil and Their Mobility in Relations to pH and Organic Carbon

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Concentrations of Pb, Zn, Cd, Ni, Cu, Cr, and Mn were determined to assess the impact of automobiles on heavy metal contamination of roadside soil. Soil samples at four polluted sites and a control site were collected at a depth of 0, 2, 5, 10, 15, 20, 30 cm. A comparison of elemental levels between polluted and control sites exhibited exceptionally higher concentrations at the former sites. The Pb levels in polluted sites varied from 70 to 280.5 $\mu g g^{-1}$ and it rapidly decreased with depth. Similarly, mean concentrations of Zn, Cd, Ni, Cu, Cr, and Mn were significantly higher at polluted sites and followed a decreasing trend with the increase in depth. Correlation coefficients between heavy metals and traffic density were positively significant except for nickel. Profile samples showed that Pb, Zn, Cd, Cu, and Mn were largely concentrated in the top 5 cm confirming airborne contamination. The vertical movement and partitioning of metals, except Ni and Cr, exhibited predominant association with soil pH and organic carbon. The results have been presented using Heavy Metal Index.

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INTRODUCTION

Soil is a precious natural resource, but its quality is deteriorated due to several anthropogenic activities. Fortunately, only rarely does soil become polluted to the extent that it can no longer support plant growth. More often, pollutants such as heavy metals are found in soil below a level toxic to plants and yet have accumulated to higher levels that may be a biohazard.

Investigations have demonstrated elevated levels of heavy metals in soils adjacent to busy roadways. The degree of heavy metal pollution, which serves as an index, is directly related to the quality of exhaust gases emanating from the combustion of leaded gasoline and the wear and tear of automobile components. Lead is a primary pollutant produced by the combustion of leaded petrol (Page and Gange, 1970), while higher levels of Zn, Cd, and Ni (Lagerwerff and Specht, 1970) and Cu and Cr were also reported (Beavington, 1973; Keyser *et al*., 1978). Soil is a dynamic and complex system where any changes in its physico-chemical characteristics would severely alter the fate of heavy metals within its body. Among the various parameters, pH and organic carbon are construed as of primary importance.

Bangalore, the fastest growing Asian city with high commercial activities, harbors over 1.5 million vehicles. The present exploratory program was to determine heavy metal contamination of roadside soil due to automobiles and their fate within the soil environment.

MATERIALS AND METHODS

Sampling

The study was carried out for 2 years covering all the seasons. The sites selected for the present study include Jnanabharathi campus — a Control site (S I), Vijayanagar (S II), Bangalore Bus Station (S III), M.G. Road (S IV) and Yeshwanthapura (S V) having the daily traffic densities of around <300, 21500, 30000, 39000, and 48000 vehicles, respectively.

Twelve soil samples were collected (0 to 2 cm) from each site at random to cover the entire sampling area. Large stones and plant materials were removed, and the samples were dried for 3 days at 60˚C and sieved through #60 nylon mesh. To determine the mobility of metals in relation to the pH and organic carbon (O.C.), 10 core samples were collected between 0 to 2, 2 to 5, 5 to 10, 10 to 15, 15 to 20, 20 to 30 cm depths using a core extractor of 2.5 cm diameter. Composite samples of respective depths were prepared, air dried for 3 days, and sieved through a 1-mm nylon mesh.

Heavy Metal Analysis

Digestion of a 0.1-g sample was carried out with 10 cm^3 of 1:1 mixture of concentrated nitric acid and hydrofloric acid contained in 50 cm^3 polypropylene squat beakers (Ward *et al*., 1975). Solutions were taken to near dryness over a waterbath and the residues were redissolved in 2 *M* hydrochloric acid, filtered through Whatman #42 filter paper, and washed repeatedly in glass distilled water to a final volume of 25 ml. Solutions were fed to Atomic Absorption Spectrophotometer (Varian Techron AA30) using acidified aqueous standards. All samples were run in triplicates.

pH and Organic Carbon

The pH of the soil was determined in soil suspensions (soil : water, 1 : 5 w/v) using Systronics–361 pH meter, while the organic carbon was estimated by the titrimetric method of Walkley and Black (1934).

Statistical Analysis

For the surface soil, Heavy Metal Index (HMI) was determined, which is the mathematical function for indicating total heavy metal pollution of each site, according to the summation equation proposed by Herzig (1993).

$$\text{Heavy Metal Index} = \sum_{i=1}^{n} \mathbf{L}\,\mathbf{C_i}$$

where LC_i represents load class that is the load category value of the i^{th} heavy metal and 'n' is the number of heavy metals used for computing HMI.

The numerical load class is hard to pick up at a glance and hence converted into dots representing proportionally increasing load representative supply (LRS). The LRS values for each metal were obtained by dividing their range values into six equidistant categories. This graphic conversion of heavy metal pollution into a pictograph enables a quick overview and allows simple direct comparisons of elements amongst the study sites. Similarly, a six-level verbal evaluation called the "predicate" of the load representative supply (LRSP) is used to facilitate the verbal evaluation. Furthermore, HMI is divided into six equidistant load categories and provided with pictographs and predicates which assist in the characterization of study sites from least to the heaviest pollution levels (Table 1).

TABLE 1
Heavy Metal Index (HMI) Categories, Pictographs and Predicates

HMI Load Categories	Pictographs	Predicates
< 7.0	•	Very low
7.01 – 14.00	•	Low
14.01 – 21.00	•	Medium
21.01 – 28.00	•	High
28.01 – 35.00	•	Very high
> 35.0	•	Critically high

RESULTS AND DISCUSSION

Surface Soil

The concentrations of the Pb across the polluted sites were higher and varied from 70 to 280.5 $\mu g g^{-1}$ compared with the control values of 2 to 3 $\mu g g^{-1}$. Mean maximum concentrations of Zn (176.4), Cd (21.1), Ni (25.5), Cu (32.3), Cr (45.5), and Mn (205.8 $\mu g g^{-1}$) also remained higher at polluted sites (Table 2).

The combustion of alkyl derivatives in gasoline by motor vehicles has contributed to extensive Pb contamination of roadside soil (Canon and Bowles, 1962; Madany *et al*., 1994; Yaman, 1995; Ramakrishnaiah and Somashekar, 1997). In the surface soil of Mission Peninsula, a light dirt road with an average daily traffic of < 50 v day^{-1}, Pb levels were < 1 $\mu g g^{-1}$ (Francek, 1997), which is comparable to the mean Pb level (2.28 $\mu g g^{-1}$) recorded for control site having < 300 v day^{-1}. Goldsmith *et al*. (1976) found soil Pb of 87.3 $\mu g g^{-1}$, at 6 m from the road carrying 21040 v day^{-1} while S II with nearly the same traffic density showed Pb ranging from 70 to 128.6 $\mu g g^{-1}$. S IV with less traffic (39000 v day^{-1}) than S V showed higher mean Pb of 270.9 $\mu g g^{-1}$. This is attributable to the long-term usage by petrol-driven vehicles. Furthermore, wide variations in Pb concentration of roadside soils ranging from 14 to 6190 $\mu g g^{-1}$ have been reported (Ward *et al*., 1977; Muskett and Jones, 1980; Harrison *et al*., 1981; Ndiokwere, 1984). These values are affected greatly by distances from the road and therefore are not directly comparable. In the present study, polluted sites with a traffic load of 21,500 to 48,000 per day possessed mean Pb of 70.2 to 270.9 $\mu g g^{-1}$, a range less than that recorded (60 to 3520 $\mu g g^{-1}$, 15,000 – > 35,000 v day^{-1}) by Yassoglou *et al*. (1987).

TABLE 2
Mean Heavy Metal Concentrations in Surface Soil ($\mu g g^{-1}$) and Heavy Metal Index of the Study Sites

Sites	Metals	Mean	± SD	n	LRS	Pictograph	LRSP	Sum	HMI
S I	Lead	2.28	0.13	24	1.00	•	Very low		
	Zinc	22.5	1.68	24	1.00	•	Very low		
	Cadmium	1.04	0.11	24	2.00	•	Low		
	Nickel	0.56	0.08	24	2.00	•	Low		
	Copper	2.08	0.61	24	2.00	•	Low		
	Chromium	1.75	0.71	24	2.00	•	Low		
	Manganese	85.24	14.67	24	2.00	•	Low	12	Low
S II	Lead	86.93	18.59	24	2.75	•	Low		
	Zinc	122.09	13.10	24	3.50	•	Medium		
	Cadmium	6.67	2.35	24	2.63	•	Low		
	Nickel	25.49	4.11	24	5.50	●	Very high		
	Copper	23.07	14.35	24	3.63	•	Medium		
	Chromium	45.49	11.62	24	4.25	●	High		
	Manganese	128.58	24.07	24	2.88	•	Low	25	High
S III	Lead	149.42	26.90	24	3.50	•	Medium		
	Zinc	144.30	10.12	24	4.00	●	High		
	Cadmium	8.41	2.70	24	3.38	•	Medium		
	Nickel	11.98	2.16	24	3.38	•	Medium		
	Copper	32.29	3.21	24	3.00	•	Medium		
	Chromium	33.89	4.41	24	4.00	●	High		
	Manganese	205.80	20.53	24	4.63	●	High	26	High
S IV	Lead	270.90	9.41	24	5.00	●	Very high		
	Zinc	158.30	8.80	24	4.13	●	High		
	Cadmium	16.17	1.47	24	4.38	●	High		
	Nickel	8.20	1.88	24	3.13	•	Medium		
	Copper	24.45	7.00	24	3.50	•	Medium		
	Chromium	33.12	11.14	24	3.63	•	Medium		
	Manganese	177.46	5.48	24	4.00	●	High	28	High
S V	Lead	202.87	13.90	24	4.25	●	High		
	Zinc	176.46	10.40	24	4.63	●	High		
	Cadmium	21.16	1.01	24	5.38	●	Very high		
	Nickel	16.05	1.39	24	4.00	●	High		
	Copper	25.35	4.92	24	3.24	•	Medium		
	Chromium	34.23	15.09	24	3.75	•	Medium		
	Manganese	175.08	9.25	24	4.00	●	High	29	Very high

In a survey of Rhondda Fawr, Burton and John (1977) observed no direct correlation despite widespread Pb in the urban strip. However, in the present case the coefficients of correlation between Pb and traffic density remained highly significant ($r = 0.906$, $P \leq 0.001$). This supports findings elsewhere that soil Pb levels increase with increase in traffic density (Yassoglou *et al*., 1987; Francek, 1997; Bharadwaj and Abhimanyu, 1998).

Across the polluted sites, the concentration of Zn, Cd, and Ni varied in the range of 101.9 to 192.2; 4.5 to 22.62; 7 to 30 $\mu g g^{-1}$, with the mean maxima of 176.4, 21.1, and 25.5 $\mu g g^{-1}$, respectively. These levels are exceptionally higher than the controls. Lagerwerff and Specht (1970) observed 114 to 162 $\mu g g^{-1}$ Zn, 0.24 to 0.94 $\mu g g^{-1}$ Cd, and 2.4 to 7.4 $\mu g g^{-1}$ Ni in the surface soil at a distance of 8 to 32 m from the road carrying 48000 v day^{-1}. Incidentally, S V with the same traffic load showed 162 to 192.2, 19.3 to 22, and 14.6 to 18 $\mu g g^{-1}$ Zn, Cd, and Ni, respectively, since sampling in the present study was much closer to the road (< 10 ft).

The major source of Zn and Cd are metal processing or impurities in Zn-based oil and rubber additives from tire wear (Lagerwerff, 1971; Haghiri, 1973; Palm and Ostlund, 1996). Beavington (1973) showed marked accumulation of Zn and Cd in the roadside soils, while Tuba and Csintalan (1993) found Ni contamination. Our study revealed significant positive correlations for Zn and Cd with traffic density (Zn, 0.958 ; Cd, 0.925 ; $P \leq 0.001$). Ward *et al*. (1977) observed significant positive correlation between soil Ni and traffic density, while in our case, although Ni levels were higher than control, the '*r*' value remained insignificant (0.386, $P \leq 0.1$). Therefore, it is concluded that enhanced levels of Zn and Cd are due to pollution by automobiles, while that of Ni is related to the texture and origin of the soil parent material as reported by Yassoglou *et al*. (1987).

Concentration of Cu, Cr, and Mn among the polluted sites increased manifoldly over the control and varied in the range, 14.2 to 60, 22.2 to 72.5 and 104.3 to 252.3 $\mu g g^{-1}$ respectively. The highest concentrations of Cu and Mn recorded at S III appear to have originated from poorly maintained city transport buses, especially during minor mechanical repairs undertaken at this site. The correlation coefficient of Cu and Cr with traffic density remained positively significant (Cu, 0.766; Cr, 0.572 ; $P \leq 0.001$), as observed by Tuba and Csintalan (1993) through transplant experiments in Hungary. A strong correlation between Mn and traffic density ($r = 0.758$, $P \leq 0.001$) constitutes an interesting result, as Lytle *et al*. (1994, 1995) suggested that the accumulation of Mn along roadways as a possible indicator of vehicular pollution.

The quality guidelines for soil heavy metal concentrations developed in certain countries indicate wide variations. In France, the soil threshold levels of heavy metals are Pb, 100; Zn, 300; Cd, 0.07; Ni, 50 and Cu, 100 mg kg^{-1} (Dachaufour, 1998). The mean and range of total heavy metal concentrations in normal soils reported by London (1984) were Pb, 35(2 to 300); Zn, 90 (1 to 900); Cd, 0.35 (0.01 to 2); Ni, 50(2 to 750); Cu, 30(2 to 250); Cr, 70(5 to 1500) and Mn, 1000 (20 to 10,000) $mgkg^{-1}$. Furthermore, limits for the total concentrations of heavy metals in

soil according to EEC (1986) were Cd, 3; Cu, 200; Pb, 200 and Zn, 300 $mgkg^{-1}$, while the Dutch reference values (VROM, 1994) are 0.8, 36, 35, and 140 $mgkg^{-1}$, respectively. Singh and Saha (1997) in their all-India coordinated research project reported Pb, 4.2 to 60; Zn, 0.8 to 119.6; Cd, 0.04 to 0.96; Ni, 0.88 to 6.78; Cu, 1.18 to 70.2; Cr, 2.1 to 9.16 and Mn, 56.6 to 59.3 $mgkg^{-1}$ soil. The critical limits used for classifying sites as toxic were Pb > 1, Zn > 2, Cd > 1, Ni > 3, Cu > 1, Cr > 5, Mn > 5 and Fe > 5 $mgkg^{-1}$. The background heavy metal concentrations in the surface soil around Bangalore recorded by Nair (1999) revealed Pb, 22.5; Zn, 57; Cd, not detected; Ni, 25; Cu, 12 and Cr, 30 $mgkg^{-1}$. This indicates that the results from our studies for Pb, Zn, Cd, Cu, and Cr reflect values greater than the natural background concentrations, which is typical of a contaminated site.

Heavy Metal Index

The heavy metal load patterns between control and polluted sites are highly comparable because they portray 'low' to 'very high' HMI categories (Table 2). The control site with a total of 12 LRS is categorized as 'Low' HMI, strongly indicating its unpolluted nature. Of the four polluted sites S II, III, and IV reached 'High' levels with the total LRS of 25, 26, and 28, respectively. Lead is classified as 'very high' at S IV with a mean of 270.9 μgg^{-1}, while Mn, Zn, and Cr were delegated to 'High' LRSP at S III. Several elements at S V reached 'High' level, namely, Pb, Zn, Ni, and Mn, and even 'Very High' in case of Cd. The massive loads measured here are attributed to heavy traffic density.

Heavy Metals in Soil Profile

Higher concentrations of metals, except Ni, were present in the top 5 cm, and thereafter decreased with depth (Table 3). Lead localization down to 15 cm has been well documented (Marsh and Siccama, 1997), while Yassaglou *et al*. (1987) found enhanced Pb down to 160 cm. In our study, considerable levels (10.75 to 31.64 μgg^{-1}) were noted down to 30 cm, which represents 8.8 to 17.2% Pb of the surface layer, indicating subsurface accumulation. This result is in accordance with the findings of Francek (1997).

There are conflicting data in the literature concerning the levels of Zn, Cd, and Cu along with the vertical gradient of the soil. By artificial contamination Khan and Frankland (1983) observed no significant Cd beyond 10 cm, while Beavington (1973) recorded predominant Zn, Cd, and Cu in the top 15 cm. Contrarily, Marsh and Siccama (1997) noted increase in Zn and Cu from 0 to 2 cm to 6 to 8 cm depth. Our data, however, consistently demonstrated maximum Zn, Cd, Cu, and Mn in the top 5 cm and decreased with increase in depth showing 41, 15.7, 21.2, and 41.6% of their respective surface levels in 20 to 30 cm. This suggests subsurface perco-

TABLE 3
Mean Heavy Metal Concentrations ($\mu g g^{-1}$), pH and Organic Carbon (%) Along the Soil Profile

Site	Depth (cm)	pH	O.C.	Pb	Zn	Cd	Ni	Cu	Cr	Mn
S I	0-2	6.43	1.28	2.25	23.7	1.16	0.55	2.82	1.50	95.40
	2-5	6.16	1.42	2.36	25.5	1.42	0.50	3.37	1.25	95.57
	5-10	5.90	1.10	1.66	22.5	0.77	0.45	1.75	0.90	71.00
	10-15	5.51	1.07	1.50	20.4	0.62	0.55	1.24	0.75	59.18
	15-20	5.40	0.97	1.00	16.4	0.50	0.72	1.28	0.62	40.30
	20-30	5.80	0.88	1.00	11.1	0.25	0.25	0.75	0.62	28.10
S II	0-2	6.88	2.02	75.3	126.8	6.70	29.25	15.75	59.50	122.80
	2-5	6.65	2.09	69.4	106.6	4.20	19.17	12.43	44.95	138.22
	5-10	6.61	1.70	40.9	95.4	3.82	20.75	7.87	51.62	106.50
	10-15	6.56	1.45	31.3	91.0	2.96	17.59	5.98	57.40	87.97
	15-20	6.50	0.80	21.9	66.0	2.07	16.50	3.97	56.92	61.21
	20-30	6.11	0.67	10.9	32.4	1.50	18.05	3.07	58.40	42.30
S III	0-2	6.68	2.01	179.50	155.88	11.72	14.20	33.50	39.00	188.12
	2-5	6.37	2.20	183.10	142.00	9.95	16.50	36.03	42.25	185.35
	5-10	6.65	1.82	167.85	156.81	8.67	15.25	23.15	43.71	147.95
	10-15	6.00	1.29	92.60	132.29	4.82	19.07	15.90	30.75	125.19
	15-20	6.03	1.09	66.75	76.33	2.20	17.32	13.57	26.55	107.37
	20-30	5.80	0.97	31.02	83.80	3.03	12.54	8.95	42.35	94.80
S IV	0-2	6.27	1.86	276.25	158.12	16.95	8.50	19.76	44.37	177.68
	2-5	6.30	1.49	260.00	161.07	16.25	10.95	12.18	39.55	148.40
	5-10	6.17	1.10	181.80	142.10	10.85	9.81	7.05	44.12	94.63
	10-15	6.12	0.89	105.01	134.20	7.10	11.40	5.65	44.67	89.70
	15-20	6.00	0.90	60.70	120.70	4.08	20.00	4.88	38.08	85.40
	20-30	5.80	0.80	28.91	107.35	1.11	15.20	5.78	39.74	75.70
S V	0-2	6.43	1.75	200.67	167.50	21.50	16.50	25.6	25.60	183.65
	2-5	6.46	2.17	205.00	183.15	16.20	19.67	37.00	34.08	187.40
	5-10	6.19	2.12	165.07	125.88	10.61	24.50	30.65	27.56	166.15
	10-15	5.92	1.50	94.58	101.37	7.03	27.50	20.75	23.00	139.35
	15-20	5.85	0.88	39.82	80.63	3.92	27.40	11.20	22.50	104.62
	20-30	5.55	0.60	17.72	61.17	3.20	28.70	6.19	19.65	93.92

lation and differential mobility within the soil body. Maximum levels of Ni and Cr in the present study were not confined to the top 5-cm layer.

Mobility of Metals in Relation to pH and Organic Carbon

The pH of the soil is in the acidic range, while organic carbon varied from 0.52 to 2.3%, and both decreased with depth (Table 3). The site wise mobility of heavy metals along the soil profile in relation to pH and O.C. provided significant positive correlations ($r > 0.800$; $P \leq 0.001$)*. Levels of Pb, Zn, and Cd also showed significant correlations, with the exception of Pb, Zn, and Cd with pH at S I, and Cd with O.C. at S V. Copper exhibited a similar relationship with O.C. at all the sites, while the trend with pH was evident at S V only. Furthermore, nickel and chromium, with the exception of with O.C. at S I and S V, did not indicate the same level of significance. The relationship between manganese and O.C. in the soil profile was found to be significant for all the sites, whereas with pH similar correlations were noted for S II, IV, and V. Therefore, it is evident that the mobility of Pb, Zn, Cd, and Mn are dependent on both pH and organic carbon.

Lead chelation by organic matter effectively retains Pb in soil (Sheppard and Thibault, 1992). Organic carbon content of 0.52 to 2.3% across the study sites appears to be sufficient to retain most of the Pb within 30 cm. Asami *et al*. (1995) attributed the low mobility of Pb in polluted soils to their affinity toward organic carbon, while Marsh and Siccama (1997) found a decrease in Pb with depth consistently correlating with organic carbon. A significant positive correlation ($P \leq 0.001$) in our case further substantiates this view (Table 4). Similarly, concentrations of Zn, Cd, Cu, and Mn strongly correlated with organic carbon ($P \leq 0.001$).

Nandram and Verloo (1985) showed low solubility of Pb, Zn, Cd, and Cu at pH 6 to 6.5 and an increase by several orders at pH 2. Similarly, Pb, Cd, and Zn exhibited weak solubilities at slightly alkaline condition (pH 8), while at pH 3.3 solubility is higher (Chaun *et al*., 1996). Furthermore with regard to Cu, Brun *et al*. (1998) reported decrease in extractable Cu with an increase in soil pH. In light of the above, the near neutral pH in our case perhaps facilitated more complexation of heavy metals with organic carbon, resulting in their accumulation in the top layers. Except for Ni, all the metals showed positive correlations with pH substantiating that the higher the pH, the more the metal retention is and *vice versa*. Although the mobility of Pb, Zn, Cd, and Mn showed significant positive correlations with pH and O.C., the trends were not uniform among all the sites. This suggests that in natural soils the mobility of metals may also depend on other soil factors in addition to pH and organic carbon.

A comparison of heavy metal content among the study sites strongly implicate automobiles as the source of contamination. Heavy metal contamination in road-

* Detailed correlation matrix for all the sites not provided for brevity.

TABLE 4
Correlation Coefficients (*r*) between Soil Characteristics and Heavy Metals Along the Soil Profile

	pH	O.C.	Pb	Zn	Cd	Ni	Cu	Cr
O.C.	0.648**							
Lead	0.381*	0.631**						
Zinc	0.497**	0.602**	0.874**					
Cadmium	0.390*	0.612**	0.933**	0.939**				
Nickel	0.159	-0.002	0.044	0.254	0.085			
Copper	0.462**	0.770**	0.640**	0.689**	0.643**	0.266		
Chromium	0.616**	0.285	0.290	0.487**	0.281	0.461**	0.353*	
Manganese	0.497**	0.832**	0.828**	0.815**	0.824**	0.192	0.801**	0.289

Significance level; * $P < 0.01$, ** $P < 0.001$.

side soil is continuous and takes place on a relatively long term basis since many metals are not so mobile. This, coupled with favorable pH and organic carbon, account for higher concentrations in the surface layers. The surface concentration of nickel and their vertical distribution in soil profile, however, remain unrelated to airborne contamination. The levels of Pb, Zn, Cd, Cu, and Cr in the roadside soil are much higher than the background levels reported for the Bangalore region. Therefore, it is apparent that this continued loading will ultimately place human health and other environmental targets at risk. In the absence of soil heavy metal regulatory limits in India, there is an urgent need to formulate threshold levels and to initiate a heavy metal pollution abatement program.

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