

Assessment of heavy metal contamination of road dusts from industrial areas of Hyderabad, India

Ramavati Mathur • V. Balaram • M. Satyanarayanan • S. S. Sawant

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Abstract Road dust in industrial areas carries high levels of toxic heavy metals. Exposure to such polluted dust significantly affects the health of people residing in these areas, which is of major concern. The present study was taken up with an aim to highlight the magnitude and potential sources of accumulation of heavy metals in 32 road dust samples collected from six industrial areas of Hyderabad. Acid-digested sample solutions were analyzed by ICP-MS for Cu, Zn, Cr, Co, Pb, Ni, V, Zr, Ce, Y, and Hf. The road dusts exhibit significantly high mean metal levels which are much above their crustal abundances. The relative ordering of mean metal contents is Zr > Zn > Pb > Cr > Ce > Cu > V > Ni > Y > Co > Hf.Elevated pollution indices (I_{geo} , EF, C_{f}^{i} , and C_{deg}) reveal that the road dusts are pollution impacted showing varying degree of heavy metal contamination. Strong positive correlations exhibited by metal pairs Cu-Zn, Cr-Ni, Ce-V, Y-Ce, and Hf-Zr imply their origin from common anthropogenic sources. Principal component analysis grouped the metals according to the sources which contributed to their accumulation. The present study confirms to an intensive anthropogenic impact on the accumulation of heavy metals in the studied road dusts attributable mainly to strong influences of vehicular and industrial activity and partly to domestic and natural processes. The results obtained imply the need for further

R. Mathur (\boxtimes) · V. Balaram · M. Satyanarayanan · S. S. Sawant

CSIR-National Geophysical Research Institute, Uppal Road, Hyderabad 500007, India

e-mail: mathurramavati@yahoo.com

investigations to assess their ecological implications and human health risks.

Keywords Road dusts · Hyderabad · Heavy metal pollution · Anthropogenic activity · Contamination assessment · Statistical analysis

Introduction

Road dust that accumulates along the impervious roadways of the urban environment acts as a sink for several pollutants especially toxic heavy metals that originate from natural geochemical processes and a vast array of anthropogenic sources, associated with vehicular traffic, industrial emissions, and domestic and commercial activity (Brown et al. 1990; Pacyna et al. 2007). These heavy metals in road dusts enter the soil, from where they get into the living tissues of plants and animals and subsequently enter the food chain of humans affecting their health and well-being and hence they have been coined as "Chemical time bombs" (Stigliani et al. 1991; Alloway and Ayres 1997; Adriano 2001). Ingestion, inhalation, or dermal contact of heavy metal-laden polluted dust leads to kidney and liver damage, different types of cancers, respiratory, neurological, cardiovascular, and skin disorders (Fergusson 1990; WHO 2007). Heavy metal pollution of urban dusts is one of the fastest growing types of environmental pollution, and it has raised serious concerns in the last decades. Extensive research carried out in the developed countries has provided important information on the content, source, accumulation, and distribution patterns of heavy metals in different segments of the environment (Charlesworth et al. 2003; Wei et al. 2010; Shi et al. 2013). However, such information from developing countries like India is scarce and is focused mainly on the heavy metal contamination of urban soils, aerosols, sediments, and water, and very few studies pertain to the same in road dusts, especially those from industrial areas (Banerjee 2003; Krishna and Govil 2008; Singh 2011; Reddy et al. 2012; Balaram et al. 2013).

Unregulated industrial activity adversely affects environmental quality which is detrimental to the health of people residing in these industrial areas and neighboring human settlements (Brown et al. 1990; Fergusson 1990). In the past decades, extensive development of industrial establishments in and around the city of Hyderabad has become a subject of concern as the industrial operations are carried out without strict environmental regulations and there is an increasing need to have an in-depth understanding of its environmental quality. Though studies had been carried out to monitor the levels, distribution and behavior of different pollutants especially heavy metals in soils, sediments and water bodies of these industrial areas (Govil et al. 2008; Krishna et al. 2009), research had not been focused on road dusts of these areas. In view of this the present work, comprising the study of 11 heavy metals in road dust samples collected from six of these severely polluted industrial areas of Hyderabad, was taken up.

The main objectives of this work are as follows: (i) to assess the concentration levels and distribution of heavy metals in the road dusts; (ii) to identify their sources using statistical tools, (iii) to evaluate the magnitude of heavy metal contamination using various indices like index of geoaccumulation (I_{geo}), enrichment factor (EF), contamination factor (C_{f}^{i}), and degree of contamination (C_{deg}); and (iv) to provide comprehensive information about the heavy metal pollution of the study area which could be useful for the authorities to plan remediation and protection approaches to achieve better environmental quality.

Materials and methods

Study area

Hyderabad, the joint capital of southern Indian states of Telangana and Andhra Pradesh, is located at $17^{\circ} 23' \text{ N}$

and 78° 28' E. It is situated on the Deccan Plateau, on an Archean granite terrain, at an average altitude of 542 m. It is one of the largest metropolitan areas of India, spreading over 650 km² and currently home to a population of 11.7 million people (7.7 million, 2011 census) (www.indiaonlinepages.com/population/hyderabadpopulation.html). The climate of the region is semi-arid with temperature varying from 20° to 39 °C, wind speed ranging from 0.2 to 9.5 km/h, and average annual rainfall of ~700 mm, mainly due to the southwest monsoon (Balakrishna and Raghava Rao 1961; Gnaneshwar and Sitaramayya 1998). In recent decades, the city has undergone rapid expansion and a tremendous increase in motorized transport resulting in frequent road congestions and a sharp increase of vehicular emissions. Many industrial areas have come up in and around the city which comprise units manufacturing different products like chemicals, pharmaceuticals, pesticides, batteries, textiles, plastic and rubber products, and metallic goods besides housing incineration plants, electroplating, tanning, and servicing workshops. As a consequence of rapid urbanization and increased economic and uncontrolled industrial development, Hyderabad is experiencing a declining trend of its environmental quality at an alarming rate (IES 2008).

Sample collection and analysis

A total of 32 road dust samples were collected from six industrial areas in and around the city of Hyderabad, namely the following: Balanagar, Sanathnagar, Jeedimetla, Patancheru, Nacharam, and Katedan (Fig. 1), details of which are given in Table 1. At each sampling site, 500 g to 1 kg (fresh weight) of composite sample was collected by thoroughly combining five sub-samples taken along the road edges on either side. Samples were collected with a plastic scoop and dustpan, wearing clean latex gloves and placed in clean labeled, self-locking polythene bags taking care to collect all the fine dust with a brush. They were air-dried for 5 days and sieved at 2 mm to remove any extraneous materials like stones and other debris and again sieved through a sieve shaker (Fritsch, Germany), and the very fine fraction (<75 µm) was homogenized in an agate mortar, oven-dried at 70 °C for 12 h before analysis. The finer fraction of the dust samples was studied because earlier studies which focused on heavy metals in different size fractions showed that many metals generally tend to be associated with and are concentrated in the



Fig. 1 Map of Hyderabad showing sampling locations

finer particle sizes and pose a higher health risk than larger particles (Lin et al. 2005). Sample solutions were prepared by closed vessel acid digestion according to the procedure given in Roy et al. (2007) using Savillex pressure decomposition vessels (M/s Savillex Corporation, Minnetonka, MN, USA), analytical reagent grade chemicals, distilled acids, and high purity water (18 M Ω). Rh was used as an internal standard. International certified reference materials (CRMs) SO-1, SO-2, and JG-2, prepared in an identical manner, were used for quality control validation (Table 2).

An ICP-MS Perkin-Elmer, Sciex (Model Elan DRC II, Toronto, Canada) was used for sample analysis to obtain data of 11 heavy metals viz.: Cu, Zn, Cr, Co, Pb, Ni, V, Zr, Ce, Y, and Hf. Scandium concentrations also obtained for the samples (Table 1) were used to get

Table 1 Concentration of heavy metals ($\mu g/g$) in road dust samples from industrial areas of Hyderabad and reference values ($\mu g/g$) (Taylorand McLennan 1985)

Samp.No	Location	Cu	Zn	Cr	Co	Pb	Ni	V	Zr	Ce	Y	Hf	Sc
RM-I-1	Balanagar	165.3	683.5	264.2	27.3	256.4	50.6	79.1	460.2	75.6	26.8	8.1	6.5
RM-I-2	Balanagar	84.3	348.8	139.8	10.1	137.8	21.6	51.7	561.5	81.5	23.1	8.8	5.6
RM-I-3	Balanagar	208.4	528.4	174.5	35.2	239.6	60.5	101.3	482.7	101.6	29.1	7.6	6.2
RM-I-4	Balanagar	56.8	152.1	168.6	16.8	160.8	58.5	104.4	530.2	124.2	24.6	6.8	5.9
RM-I-5	Balanagar	269.2	652.3	225.9	20.8	299.3	54.4	70.6	598.1	90.2	32.4	7.3	7.3
RM-I-6	Sanathnagar	276.2	990.2	242.6	29.7	288.8	83.0	147.3	548.5	149.8	28.3	7.8	7.8
RM-I-7	Sanathnagar	62.3	198.3	60.4	8.2	104.6	22.4	70.9	706.9	105.7	23.7	8.6	6.8
RM-I-8	Sanathnagar	162.7	244.5	99.6	8.5	127.5	49.7	51.5	297.5	91.9	24.4	8.4	6.4
RM-I-9	Sanathnagar	322.2	734.6	194.2	14.8	269.5	79.3	81.6	313.4	172.6	31.5	8.8	8.8
RM-I-10	Sanathnagar	76.6	226.3	115.3	10.4	132.5	56.8	61.3	276.2	120.8	29.2	7.1	7.1
RM-I-11	Jeedimetla	121.6	482.0	142.9	10.9	164.6	40.9	57.0	770.4	104.6	28.2	12.7	7.3
RM-I-12	Jeedimetla	34.1	131.3	82.2	17.3	99.3	39.1	83.5	539.0	147.6	29.3	8.8	10.7
RM-I-13	Jeedimetla	45.2	121.3	75.1	4.8	82.1	25.8	68.1	753.8	74.1	19.8	11.0	5.2
RM-I-14	Jeedimetla	36.7	101.3	72.7	5.4	68.2	47.5	34.8	300.7	85.3	21.6	5.1	4.4
RM-I-15	Jeedimetla	101.7	515.9	169.2	15.0	227.4	53.9	73.5	475.3	135.7	32.2	10.4	6.8
RM-I-16	Patancheru	201.1	382.8	228.9	23.1	62.3	98.5	58.0	495.8	78.3	13.2	4.3	6.4
RM-I-17	Patancheru	146.9	235.6	250.7	17.3	121.2	80.9	60.9	636.7	132.1	31.2	13.0	6.9
RM-I-18	Patancheru	215.1	325.2	158.3	14.9	126.6	115.3	56.7	372.6	64.0	11.2	6.1	5.7
RM-I-19	Patancheru	127.7	216.3	357.9	38.9	68.4	97.4	104.7	304.5	55.8	16.8	3.5	7.2
RM-I-20	Patancheru	41.9	93.7	74.9	16.9	62.4	47.7	82.4	425.1	83.1	20.7	3.9	4.4
RM-I-21	Patancheru	58.2	110.0	98.6	16.0	57.8	39.5	89.0	493.9	97.5	21.7	5.8	9.9
RM-I-22	Patancheru	54.9	82.4	122.4	25.1	58.8	76.2	128.2	432.2	211.1	33.7	4.2	7.6
RM-I-23	Nacharam	148.5	717.6	103.8	19.6	139.7	37.3	101.0	687.7	168.8	41.5	13.2	6.9
RM-I-24	Nacharam	38.3	113.1	96.8	22.3	48.7	15.2	169.5	560.2	284.8	52.8	7.7	10.2
RM-I-25	Nacharam	45.8	190.4	72.4	4.8	80.5	28.8	64.1	767.5	114.3	49.3	10.2	5.7
RM-I-26	Nacharam	56.9	240.1	79.2	5.2	112.7	35.3	80.7	527.6	156.9	37.1	8.5	6.3
RM-I-27	Katedan	179.9	439.1	104.7	8.3	589.1	41.9	82.0	1085.8	117.4	49.9	21.0	10.5
RM-I-28	Katedan	93.3	146.7	86.8	23.4	89.5	46.5	101.4	156.3	189.7	28.7	4.0	12.7
RM-I-29	Katedan	37.3	89.0	81.0	28.3	296.8	42.5	170.4	631.8	238.2	55.7	15.2	7.4
RM-I-30	Katedan	39.1	176.1	50.9	4.7	225.1	11.3	50.5	696.6	98.9	28.6	11.0	8.6
RM-I-31	Katedan	48.2	90.2	103.4	16.4	283.6	42.9	86.4	425.7	93.5	23.2	8.6	11.9
RM-I-32	Katedan	158.6	299.9	93.2	6.2	256.4	25.8	55.7	544.7	112.4	33.3	9.3	6.8
UCC		25.0	71.0	35.0	10.0	20.0	20.0	60.0	190.0	64.0	22.0	5.8	11.0

UCC upper continental crust (Taylor and McLennan 1985)

normalized enrichment factor. The instrumental parameters and operating conditions of the ICP-MS were set using a procedure described by Balaram and Rao (2003), such that a uniform sensitivity was obtained for all the masses across the entire mass range. The precision of the measurement was <5 % RSD. The

Table 2 Concentration of heavy metals $(\mu g/g)$ obtained in certified reference materials used for the present study

Ref. material	SO-1 ^a		SO-2 ^b		JG-2 ^c		
Analyte	This study ^d	Certified value ^e	This study ^d	Certified value ^e	This study ^d	Certified value ^e	
Cu	60.4	61	8.4	8	0.4	0.4	
Zn	141.2	144	119.2	115	12.4	12.7	
Cr	167.4	170	13.1	12.3	6.2	7.6	
Co	28.4	29	8	7.6	4.3	4.3	
Pb	19.6	20	21.1	20	31.7	32.8	
Ni	89.8	92	8.5	8	2.1	2.1	
V	130.4	133	55.2	57	2.3	3	
Zr	85.7	84	779.2	760	100.4	101	
Ce	103.9	102	115.2	112	50.6	49.5	
Y	25.2	24.5	41.4	40	89.7	88.2	
Hf	2.4	2.5	18.4	17.4	5.3	5.4	
Sc	17.2	17.7	11.1	11.3	2.4	2.5	

^a Regosolic clay soil, CCRMP, Canada

^b Podzolic B horizon soil, CCRMP, Canada

^c Biotite granite, Geological Survey of Japan, Japan

^d Average of six measurements

^e Source: Govindaraju (1994)

certified values for the international reference materials, used for external calibration, were obtained from Govindaraju (1994) (Table 2).

Data analysis

Basic statistical data

Descriptive statistical data analysis was performed using SPSS version 7.5 for Windows. The basic statistical attributes (min., max., mean, median, range, SD, kurtosis, skewness, coefficient of variation (CV)) (Table 3) were computed to have an elementary understanding of the heavy metal data and to know the activeness of the metals in the examined environment.

Contamination assessment methods

Assessment of the intensity of heavy metal pollution of the examined environment was carried out using the index of geoaccumulation, enrichment factor, contamination factor, and contamination degree, by comparing observed heavy metal concentrations to background concentrations (Loska et al. 2004). In this study, the upper continental crust (UCC) values for metals given by Taylor and McLennan (1985) (Table 1) were adopted as background values (reference values).

The Index of geoaccumulation (I_{geo}) introduced by Muller (1969) was calculated using the equation: $I_{geo} = \log_2 C_n / 1.5 B_n$

where C_n represents the measured concentration of the examined element (n) in the sample (road dust) and B_n is the background value (UCC) of the element (n). The constant 1.5 is the background matrix correction factor due to lithogenic variability (Loska et al. 2004). The descriptive classification for I_{geo} (Muller 1981) includes seven grades starting from class 0 (uncontaminated) to class 6 (extremely contaminated) as follows: $I_{geo} \leq 0$ practically uncontaminated; $0 < I_{geo} < 1$ uncontaminated to moderately contaminated; $1 < I_{geo} < 2$ moderately contaminated; $3 < I_{geo} < 4$ heavily contaminated; $4 < I_{geo} < 5$ heavily to extremely contaminated; $5 < I_{geo}$ extremely contaminated.

Enrichment factor (EF) is based on normalization of a tested element against a reference element (Al, Fe, Ti, Sc, Mn) which is not subject to contamination by anthropogenic sources (Reimann and de Caritat 2005). In this study, scandium (Sc) was chosen as the reference element. The EF value was calculated using the

Table 3 Statistical summary of heavy metals in the studied road dusts

	Cu	Zn	Cr	Co	Pb	Ni	V	Zr	Ce	Y	Hf	Sc
Mean	116.09	314.34	137.22	16.46	166.81	50.84	83.69	526.85	123.69	29.78	8.65	7.43
Median	88.80	230.95	104.25	16.20	130.00	47.00	79.90	528.90	109.05	28.65	8.45	6.90
SD	80.59	235.62	72.79	9.15	113.45	25.07	32.77	185.18	51.98	10.66	3.66	2.03
CV	0.69	0.75	0.53	0.56	0.68	0.49	0.39	0.35	0.42	0.36	0.42	0.27
Kurtosis	0.05	0.80	1.25	-0.15	4.77	0.33	1.56	1.47	2.03	0.68	2.99	0.62
Skewness	0.95	1.21	1.25	0.62	1.78	0.81	1.30	0.62	1.40	0.86	1.28	1.00
Range	288.10	907.80	307.00	34.20	540.40	104.00	135.60	929.50	229.00	44.50	17.50	8.30
Minimum	34.10	82.40	50.90	4.70	48.70	11.30	34.80	156.30	55.80	11.20	3.50	4.40
Maximum	322.20	990.20	357.90	38.90	589.10	115.30	170.40	1085.80	284.80	55.70	21.00	12.70

modified formula given by Loska et al. (2004) based on the equation suggested by Buat-Menard and Chesselet (1979).

 $\text{EF} = \left[C_n(\text{sample}) \middle/ C_{ref}(\text{sample}) \right] \middle/ \left[B_n(\text{background}) \middle/ B_{ref}(\text{background}) \right]$

where C_n (sample) is the content of the examined element in the examined environment (road dust), C_{ref} (sample) is the content of the reference element in the examined environment, B_n (background) is the content of the examined element in the reference environment (UCC), and B_{ref} (background) is the content of the reference element in the reference environment. Five contamination categories are recognized on the basis of the EF (Sutherland 2000) as follows: EF < 2 deficiency to minimal enrichment; EF = 2–5 moderate enrichment; EF = 5–20 significant enrichment; EF = 20– 40 very high enrichment; EF > 40 extremely high enrichment.

Contamination factor $(C_{\rm f}^{\rm i})$ and degree of contamination $(C_{\rm deg})$ and their respective categories were suggested by Hakanson (1980). $C_{\rm f}^{\rm i}$ is the single element index and $C_{\rm deg}$ is the sum of contamination factors for all elements examined indicating the overall contamination of the environment. The contamination factor $(C_{\rm f}^{\rm i})$ was calculated using the equation:

$$C^{i}_{f} = C^{i}_{0-1} / C^{i}_{n}$$

where C_{0-1}^{i} is the mean content of metals from at least five sampling sites and C_{n}^{i} is the pre-industrial concentration of individual metal. In the present study, modification of the factor (Loska et al. 2004) was used wherein the reference values were of UCC. C_{n}^{i} is defined according to four categories as follows: $C_{\rm f}^{i} < 1$ low contamination; $1 \le C_{\rm f}^{i} < 3$ moderate contamination; $3 \le C_{\rm f}^{i} < 6$ considerable contamination; $6 \le C_{\rm f}^{i}$ very high contamination. The contamination degree ($C_{\rm deg}$) of the environment has four classes which are as follows: $C_{\rm deg} < 8$ low degree of contamination; $8 \le C_{\rm deg} < 16$ moderate degree of contamination; $16 \le C_{\rm deg} < 32$ considerable degree of contamination; $32 \le C_{\rm deg}$ very high degree of contamination.

Statistical analysis

Pearson's correlation coefficient analysis and principal component analysis (PCA) were carried out using the software SPSS version 7.5 for Windows, to evaluate the associations of the studied heavy metals and establish the strength of their interrelationship for identifying their sources and pathways. PCA was used to extract a small number of latent factors (principal components) for analyzing relationships among the observed variables. Varimax rotation with Kaiser normalization (Kaiser 1960) was applied to maximize the variances of the factor loadings across variables for each factor for better interpretation of results.

Results and discussion

Elemental concentrations

The heavy metal data of the studied road dusts and UCC and the international certified reference materials are presented in Tables 1 and 2, respectively. Table 3 summarizes the basic statistical attributes of these road dusts. Elemental data shows a large variation (Table 1). Zn and Zr record the highest levels with values ranging from 82.4 to 990.2 µg/g and 156.3 to 1085.8 µg/g, respectively. About 38 % of the samples have Zn contents $>300 \mu g/g$, 53 % of them exhibit Zr concentrations $>500 \mu g/g$, but only six samples have Cu levels above $200 \mu g/g$ (Table 1). The mean levels of Ce (123.7 $\mu g/g$), Zr $(526.9 \,\mu\text{g/g})$, Y $(29.8 \,\mu\text{g/g})$, and Hf $(8.7 \,\mu\text{g/g})$ obtained for the present study are higher in comparison to the mean levels of these metals reported for road dusts from different traffic areas of Hyderabad (Mathur et al. 2011) which indicate that industrial activity mainly contributed to the enrichment of these metals. The ranges of the contents of the heavy metals are also quite wide. The maximum values of Cu and Cr are 9.5 and 7 times, respectively, higher than their minimum values while maximum Zn and Pb levels are 12 times greater than their minimum levels (Table 3). The relative ordering of mean contents of metals is Zr > Zn > Pb > Cr > Ce > Cu > V > Ni > Y > Co > Hf.The mean concentrations of Pb (166.8 μ g/g), Zr (526.9 μ g/ g), and Ni (50.8 µg/g) are 8.3, 2.8, and 2.5 times, respectively, higher while the mean contents of Cu, Zn, and Cr are about 4 times higher than their corresponding crustal (UCC) abundances (Tables 1 and 3). The mean concentrations of Co, V, Ce, Y, and Hf are 1.4 to 1.9 times higher than the reference values.

All the industrial sites display a wide range for most elements with samples from each area exhibiting both high as well as low values (Table 1). However, road dusts from Balanagar and Sanathnagar industrial areas are identified as having higher levels of Zn, Cu, Cr, and Pb while those from Patancheru have higher Ni, Co, and Cr contents. Samples from Jeedimetla exhibit moderate levels of most elements in comparison to other areas. Highest mean concentrations of Pb and Hf are displayed by Katedan road dusts while those from Nacharam and Katedan have higher mean concentrations of Ce, Y, Zr, and V (Table 1). Govil et al. (2008) had also reported high concentrations of Pb, Cr, Ni, and Zn in soils from the Katedan industrial area. Thus, it can be inferred that the heavy metal concentrations differ depending on the predominant activities of the sampling sites and local pollution sources. The contents of Cu and Zn obtained in the present study are lower than those reported for street dusts of Xianyang (Shi et al. 2013), Madrid (De Miguel et al. 1997), and Birmingham and Coventry (Charlesworth et al. 2003) (Table 4). The levels of most of the metals in road dusts reported by Ahmed et al. (2007), Singh (2011), and Wei et al. (2010) are either low or comparable to those of the present work. However, the values of Zn, Cr, and Pb reported by Yongming et al. (2006) for urban dusts of Xi'an, China, and Cr and Cu values for soils from Manali industrial area, Chennai, India (Krishna and Govil 2008) (Table 4) are higher than those of the present work.

The mean and median values of Co, Ni, Zr, and Y are the same (Table 3). Regardless of their skewed distribution, the mean and median values of V and Hf are also close. The coefficient of variation for Cu, Zn, and Pb is more than 0.6 in comparison to CV's of other metals (Table 3). Skewness coefficient of Zn, Cr, Pb, and Ce exceeds 1, which measures the asymmetry and indicates that these metals are positively skewed toward lower concentrations as revealed by the fact that the median concentrations of these metals are lower than their mean concentrations. Zn contents show larger variability with a range of 907.8 (82.4 to 990.2 µg/g) and SD of 235.6 of which five samples have high concentrations $>650 \mu g/g$ (Tables 1 and 3). The highest values of skewness and kurtosis are found for lead (Table 3) suggesting that its distribution is not normal and it has a broad range of concentrations characterizing a strong anthropogenic influence. Kurtosis calculation of data shows negative value for Co implying a more even distribution of this metal while positive values of most heavy metals (Table 3) indicate a peaked distribution with sporadic high emissions of these elements from certain sources.

Assessment of pollution levels

Determination of the index of geoaccumulation, the enrichment factor, and contamination factor helped for the quantification of heavy metal accumulation in the studied road dusts from Hyderabad. The contamination degree of the dusts was useful in assessing the overall pollution level of metals in the study area.

Index of geoaccumulation

The box plot of I_{geo} for metals is displayed in Fig. 2. Based on I_{geo} results, the road dusts can be categorized as practically uncontaminated to moderately contaminated with respect to the studied heavy metals except Pb. Y exhibits the lowest mean I_{geo} value followed by V as most of the samples have $I_{\text{geo}} < 0$ for these metals (Fig. 2). None of the samples have I_{geo} values >1 for V and Y. With respect to Co, V, Y, and Hf, these dusts are practically uncontaminated with mean $I_{\text{geo}} < 0$ while Ni,

Location	Cu	Zn	Cr	Co	Pb	Ni	V	Zr	Ce	Y	Hf	Reference
Hyderabad	116.09	314.34	137.22	16.46	166.81	50.84	83.69	526.85	123.69	29.78	8.65	Present work
Dhaka	104.9	168.7	136.1	-	53.3	35.2	-	210.6	76– 103	-	9.0– 13.6	Ahmed et al. (2007)
Xianyang	132.17	375.38	135.63	-	77.3	69.54	_	-	-	-	_	Shi et al. (2013)
Urumqi	100.81	349.07	55.86	10.97	57.03	43.54	_	-	-	_	_	Wei et al. (2010)
Xi'an	94.98	421.46	167.28	_	230.52	_	_	-	-	_	_	Yongming et al. (2006)
Birmingham	466.9	534	-	_	48	41.1	_	-	-	_	-	Charlesworth et al. (2003)
Coventry	226.4	385.7	-	-	47.1	129.7	-	-	-	-	-	Charlesworth et al. (2003)
Madrid	188	476	61	3	1927	44	17	_	_	_	_	De Miguel et al. (1997)
Manali, Chennai	154.4	128.2	221.7	16.9	41.8	48.9	71.7	-	-	-	-	Krishna and Govil (2008)
Dhanbad and Bokaro	26	78	57	10	48	24	57	_	_	_	_	Singh (2011)

Table 4 Comparison of heavy metal concentrations (µg/g) of road dusts between different cities

Zr, and Ce exhibit mean I_{geo} values which categorize them to be uncontaminated to moderately contaminated. I_{geo} data obtained reveals that most of the contaminant metals are considerably enriched when compared to their crustal abundances, though none of the samples exhibit I_{geo} values that indicate extreme contamination. Igeo values of dust samples for Cu, Zn, Cr, and Pb (Fig. 2) indicate that pollution due to these heavy metals is wide spread, especially samples from Balanagar and Sanathnagar industrial areas that exhibit higher values for these metals (Table 1). Road dusts from Katedan industrial area are heavily contaminated with respect to Pb. I_{geo} data reveals that high levels of toxic metals in the study area are derived predominantly due to anthropogenic activities with differential accumulation of these metals.

Enrichment factor

The distribution of each metal's EF is displayed in Fig. 3. Pb shows the highest values of EF ranging from 2.63 to 30.86 with a mean of 12.44 (Fig. 3). The EFs for Cu, Zn, and Cr range from 1.40 to 16.60, 1.17 to 19.67, and 1.86 to 15.62 with mean values of 7.21, 6.91, and 6.18, respectively (Fig. 3), and along with Pb are ranked as a group showing significant enrichment based on the classification of Sutherland (2000). The elevated EFs observed for Cu, Zn, Cr, and Pb are mainly due to significant enrichment of these metals in dust samples

from Balanagar, Sanathnagar, and Katedan. The lowest mean EF of 2.08 is obtained for Y, with values ranging from 0.97 to 4.32, followed by V which exhibits mean EF of 2.12 (Fig. 3). This is similar to the results observed for I_{geo} (Fig. 2). The minimum EFs displayed by all the metals except Pb are <2 indicating deficiency to minimal enrichment (Sutherland 2000). None of the analyzed heavy metals have mean EF showing very high enrichment (Fig. 3). Based on the mean EFs, the road dusts can be categorized as moderate to significantly enriched with respect to all the metals studied. Though the mean EFs of Ni and Zr are higher showing values of 4.05 and 4.35, respectively, when compared to the mean EFs of Co, V, Ce, Y, and Hf (Fig. 3), they all are grouped



Fig. 2 Box plot of index of geoaccumulation for heavy metals in the studied road dusts



Fig. 3 Box plot of enrichment factors for heavy metals in the studied road dusts

into the same class showing moderate enrichment, with samples from Nacharam being more enriched in Ce, Y, V, and Hf. Samples from Jeedimetla and Patancheru display moderate to significant enrichment of Cu, Zn, Cr, Pb, Ni, and Zr. The lowest mean EF found for Y implies contribution also from natural sources. According to Sutherland (2000), EF value <2 indicates deficiency to minimal enrichment implying contribution of the metal from natural processes. On the other hand, Cu, Zn, Cr, and Pb with maximum EF > 15 (Fig. 3) suggest that anthropogenic sources contributed a substantial fraction of these elements to the road dusts. The maximum EF for all the studied metals ranges from 4.01 to as high as 30.86 (Fig. 3) due to the difference in the input of each metal to the dusts reflecting varying degree of anthropogenic pollution. Maximum EF of 30.86 for Pb indicates serious contamination due to site-specific pollution sources.

Contamination factor and degree of contamination

Table 5 displays the contamination factor evaluated for the studied heavy metals in this work. Highest mean $C_{\rm f}^{\rm i}$ of 8.34 displayed by Pb implies very high contamination level of this metal according to the classification categories given by Hakanson (1980). Pb also displays highest levels for $I_{\rm gco}$ and EF. On the basis of mean $C_{\rm f}^{\rm i}$ the road dusts are classified as considerably contaminated by Cu, Zn, and Cr but moderately contaminated with respect to all other studied metals. However, maximum $C_{\rm f}^{\rm i}$ for Cu (12.89), Zn (13.95), and Cr (10.23) (Table 5) indicates very high contamination levels attributable to road dusts mainly from Balanagar and

 Table 5
 Summary of contamination factor and degree of contamination for heavy metals in the studied road dusts

Element	Min C_f^i	Max C_f^i	Mean C_f^i
Cu	1.36	12.89	4.64
Zn	1.16	13.95	4.43
Cr	1.45	10.23	3.92
Со	0.47	3.89	1.65
Pb	2.44	29.46	8.34
Ni	0.57	5.77	2.54
V	0.58	2.84	1.39
Zr	0.82	5.71	2.77
Ce	0.87	4.45	1.93
Y	0.51	2.53	1.35
Hf	0.6	3.62	1.49
C _{deg}	10.84	95.33	34.47

Sanathnagar industrial areas, as discussed earlier. Maximum C^{i}_{f} values of Co, Ni, Zr, Ce, and Hf (Table 5) indicate considerable contamination (Hakanson 1980). On the whole, C^{i}_{f} trends confirmed the results achieved applying I_{geo} and EF indices. The C_{deg} for the mean metal levels of the road dusts is 34.47 (Table 5) denoting very high degree of contamination (Hakanson 1980). Data presented in Table 5 indicates that Pb contributed most to the degree of contamination (24.19 %) followed by Cu (13.46 %), Zn (12.85 %), and Cr (11.37 %), respectively. Individual contributions of Co, V, Y, and Hf to C_{deg} were less than 5 %.

Based on the evaluated pollution indices (I_{geo} , EF, and C_{f}^{i}), comparative trends of metal pollution levels in decreasing order are as follows:

$$\begin{split} I_{\text{gco}} : \text{Pb} > \text{Cu} > \text{Cr} > \text{Zn} > \text{Xr} > \text{Ni} > \text{Ce} > \text{Co} > \text{Hf} > \text{V} > \text{Y}.\\ \text{EF} : \quad \text{Pb} > \text{Cu} > \text{Zn} > \text{Cr} > \text{Zr} > \text{Ni} > \text{Ce} > \text{Co} > \text{Hf} > \text{V} > \text{Y}.\\ \text{C}^{i}_{\text{f}} : \text{Pb} > \text{Cu} > \text{Zn} > \text{Cr} > \text{Zr} > \text{Ni} > \text{Ce} > \text{Co} > \text{Hf} > \text{V} > \text{Y}. \end{split}$$

The evaluated pollution indices reveal that road dusts from the industrial areas of Hyderabad are pollution impacted showing varying degree of heavy metal contamination.

Correlation coefficient analysis

Pearson's correlation analysis was applied to the total concentration of heavy metals to establish their interrelationship. The correlation matrix (Table 6) shows that there is a strong positive correlation (r > 0.7) between the metal pairs of Cu-Zn, Cr-Ni,

				5							
	Cu	Zn	Cr	Со	Pb	Ni	V	Zr	Ce	Y	Hf
Cu	1.00										
Zn	0.82 ^b	1.00									
Cr	0.61 ^b	0.52 ^b	1.00								
Co	0.31	0.27	0.65 ^b	1.00							
Pb	0.45 ^b	0.48 ^b	0.11	0.02	1.00						
Ni	0.57^{b}	0.29	0.71 ^b	0.52 ^b	-0.04	1.00					
V	-0.06	0.04	0.09	0.65 ^b	0.08	0.04	1.00				
Zr	-0.07	0.12	-0.21	-0.31	0.44 ^a	-0.42	0.01	1.00			
Ce	-0.16	-0.07	-0.25	0.21	-0.001	-0.19	0.75 ^b	0.01	1.00		
Y	-0.13	0.04	-0.30	-0.02	0.38 ^a	-0.43	0.50^{b}	0.47 ^b	0.73 ^b	1.00	
Hf	0.05	0.20	-0.21	-0.31	0.66 ^b	-0.35	0.02	0.79 ^b	0.17	0.61 ^b	1.00

Table 6 Pearson's correlation matrix of heavy metals in the studied road dusts

^a Correlation is significant at the 0.05 level (two-tailed)

^b Correlation is significant at the 0.01 level (two-tailed)

Ce-V, Y-Ce, and Hf-Zr, with maximum value of 0.82 displayed by Cu and Zn. Significant positive correlations (between 0.7 and 0.5) are also exhibited between Cr-Cu, Zn, Co; Ni-Cu, Co; V-Co, Y; and Hf-Pb, Y at the 0.01 level. Zr and Y also present a positive correlation. The positive correlation coefficients for these metals imply that they originated from common anthropogenic sources. Pb on the other hand displays a less pronounced positive correlation with Cu, Zn, Zr, and Y. Association of Pb with Cu and Zn as well as Hf, Zr, and Y indicates its contribution to the dusts from multiple sources since Cu and Zn unlike Pb display very low to negative correlation with Hf, Zr, and Y (Table 6). The correlation of Cu, Zn, and Cr in the dusts demonstrates that these metals are deposited from anthropogenic sources since there is no known geogenic source in the study area which can contribute to this type of association. The correlation matrix shows that there is very low to negative correlation between the other metal pairs especially of Cu, Zn, Cr, and Ni individually with V, Zr, Ce, Y, and Hf which signify that the sources for accumulation of these metals are much different and they may have accumulated through independent or multiple pathways. Since the studied heavy metals are commonly used as components of automobile parts and for different industrial processes, their accumulation in the road dusts is attributed mainly to industrial activity and vehicular emissions with minor contributions also from domestic activity and natural sources.

Principal component analysis

In the present study, PCA helped to identify significantly important variables and the possible sources of the heavy metals to the road dusts. According to the Kaiser criterion, the number of significant principal components with eigen values higher than 1 were selected. The factor loadings, eigen values of the factors, and % variances are listed in Table 7. Factor loadings >0.5 (italicized characters in Table 7) are regarded as significant in the interpretation of data. Three factors are obtained accounting for 80.49 % of the total variance (Table 7). All the elements are well represented by the three factors. Out of the total variance, 32.64 % is controlled by factor 1, showing higher loadings for Cu, Cr, Zn, Ni, and Co. Association of these metals in the group imply that they mainly originated from similar pollution sources. Correlation analysis (Table 6) also displays significant positive correlations between them which are an indicator of the same source. It is also observed that the loading of Co (0.61) is not as high as the loading of the other metals of this group implying that Co may have an additional source which contributed to its concentration. This factor suggests a source relating to both vehicular and industrial emissions (anthropogenic factor) since the elements grouped under

 Table 7
 Rotated component matrix of heavy metal data of studied road dusts

Analyte	Factor 1	Factor 2	Factor 3
Се	-0.20	0.11	0.89
Co	0.61	-0.36	0.57
Cr	0.86	-0.22	-0.01
Cu	0.89	0.19	-0.13
Hf	-0.07	0.93	0.07
Ni	0.74	-0.42	-0.04
Pb	0.39	0.77	0.04
V	0.11	-0.04	0.94
Y	-0.22	0.61	0.67
Zn	0.79	0.37	-0.03
Zr	-0.16	0.82	-0.02
Eigen value	3.59	2.96	2.30
% of variance	32.64	26.94	20.91
Cumulative %	32.64	59.58	80.49

Factor loadings >0.5 are represented as italicized characters

this factor are associated with vehicle engine exhaust, wearing, and corrosion of automobile parts besides their emission from metal working and fabrication units, chemical, and other industries (De Miguel et al. 1997; Hjortenkrans et al. 2007; Thorpe and Harrison 2008).

The variance explained through the second factor is 26.94 % of the total variance and is mainly characterized by high positive loadings of Hf (0.93), Pb (0.77), Y (0.61), and Zr (0.82) (Table 7). In this group, Y has a loading lower than that of other metals of the group while Pb exhibits a higher loading (0.77) for this factor in comparison to its less significant loading (0.39) for factor 1. The metals exhibiting high loadings for factor 2 are employed in different industrial processes (Kogel et al. 2006) and hence can be defined as anthropogenic components related mainly to industrial emissions. Zr, Y, and Hf are also components of the automobile catalytic converters (Helmers 1996; Jarvis et al. 2001) and may have been partially contributed by autocatalystderived traffic emissions. Though the metals showing significant correlation with factor 1 are also identified as traffic and industry-related metals, their lower to negative correlation with factor 2 (Table 7) implies sources which are different from those which contributed Hf, Zr, and Y to the road dusts in the study area. Results of correlation analysis also substantiate this inference as Cu, Zn, Cr, and Ni exhibit very low to negative correlation with these metals (Table 6). Correlation of Pb both with factors 1 and 2 and of Y with high loadings for factors 2 and 3 (0.61 and 0.67, respectively) indicate their association with heavy metals of these factors and release from similar sources. Factor 3 is dominated by Ce, Co, V, and Y explaining 20.91 % of total variance. The loading of Co (0.57) is lower than the other metals of this group. Rotated component matrix shows that Co displays a combined relationship with factor 1 and factor 3 indicating that it has a combined origin. It exhibits a strong correlation especially with Cr, Ni, and V but a very low to negative correlation with Ce and Y (Table 6). Von the other hand displays a strong positive correlation not only with Co but also with Ce and Y indicating its contribution from multiple sources. This factor can be assigned to a source of mixed origins with contributions not only from industrial sources but also from natural sources, vehicular, and domestic activity. Ce, Y, Co, and V all have different uses in vehicular systems and industrial processes especially in the metal processing industries and as fuel additives (Helmers 1996; Jarvis et al. 2001). In general, results of PCA agreed well with that of the correlation coefficient analvsis. Thus, PCA results indicate that different kinds of industrial activity, exhaust, and non-exhaust traffic emissions clubbed with contributions from domestic activity and less significantly from geogenic (natural) sources might have contributed to the accumulation of heavy metals in the studied road dusts.

Source identification

Results obtained in the present study indicate that anthropogenic influence on the concentration of heavy metals in the road dusts is more pronounced. Correlation coefficient analysis and PCA results (Tables 6 and 7) are quite consistent and give major information about the sources of these metals which can be assigned mainly to industrial and traffic emissions with contributions also from domestic and natural sources. Cu, Zn, Cr, Ni, and Co grouped together under factor 1 of PCA (Table 7) are strongly correlated (Table 6) and exhibit a wide range of concentrations and high mean levels in comparison to their corresponding crustal abundances (Tables 1 and 3) which imply to their origin from similar anthropogenic sources. They are used in fabrication of automobile metallic parts like body panels, steel studs, tyres, brake pads, brake lining systems and are also components of road marking paint, lubricating oil (engine oil), and fuel especially diesel fuel (Hjortenkrans et al. 2007; Pacyna et al. 2007; Thorpe and Harrison 2008; Wik and Dave 2009). Thus, these metals are emitted to the roadway environment due to both exhaust and non-exhaust vehicular emissions resulting from fuel combustion, lubricating oil spills, mechanical abrasion, and corrosion of metallic automobile parts. Zn, Cu, and Cr are part of different alloys especially steel and brass alloys while Cr, Co, and Ni are used in tanning, textiles, and manufacture of plastic products, paints, glass, and ceramics (Adriano 2001; Romic and Romic 2003; Rodriguez et al. 2004). Ni is also used in manufacture of batteries and fertilizers (Sutherland 2000). Hence, metallurgical and chemical processes, manufacture of goods, fuel combustion, panel beating, rusting of metal scrap, and waste incineration release these metals which accumulate in the roadway environment. In keeping with the observations of Banerjee (2003), Romic and Romic (2003), and Ahmed et al. (2007), it is considered that these heavy metals have originated from industrial and traffic emissions.

Pb is associated with vehicular, domestic, and industrial pollution (Parekh et al. 2002; Charlesworth et al. 2003; Wei et al. 2010) which is reflected by its positive correlation with Zn, Cu, Zr, Y, and Hf (Table 6) and loadings for factors 1 and 2 of PCA (Table 7). It is a component of brake lining material and wheel weights (Hjortenkrans et al. 2007). Lead was being used as an octane enhancer in gasoline which has been phased out (De Miguel et al. 1997). However, Pb containing petrol and diesel is still being used in vehicles at unchecked points in the study area (Mathur et al. 2011). A significant amount of Pb is employed in the manufacture of paints and dyes, pesticides, fertilizers, batteries, glass and as an impurity of Zn is present in galvanized metals (Parekh et al. 2002; Yongming et al. 2006). Its high concentration and wide range, in the studied dusts, could be a result of emissions from these multiple sources. Part of the Pb could have also accumulated from domestic activity like burning of fuel, use of Pb-based products, and Pb-painted construction material. Zr, Hf, and Y are employed as catalysts in the chemical and metallurgical industry, in electronics, leather tanning and to improve the properties of metal alloys (Schaller 1991; Kogel et al. 2006) which explains for their loading and association with factor 2 of PCA (Table 7). Hf is always associated with Zr indicating to their chemical association which explains for the observed high positive correlation (0.79) of these two metals (Table 6). Y is used in automobile oxygen sensors, in certain spark plugs and along with Ce is employed in ceramic and glass industry which could be the reason for its variation and loading in both factors 2 and 3 of PCA (Table 7). It is employed as a Pb replacement in anti-corrosion coatings, and hence, it exhibits a moderate correlation with Pb (Table 6) (Goering et al. 1991). Zr, Hf, and Y are important components of catalytic converters in automobiles and along with Pt, Pd, and Rh are used for the catalyst's wash coat (Helmers 1996; Zereini et al. 2001). Zr oxides and Zr silicates are used in brake lining materials along with Pb (Schaller 1991). But, autocatalyst-derived traffic emissions do not appear to have significantly contributed these metals in view of the fact that the type of vehicles, especially the transport vehicles, trucks, and auto rickshaws plying in these industrial areas are more than 15 years old, and most of them may not be fitted with catalytic converters. Unlike in the developed countries, catalytic converters fitted to automobiles have been introduced very late in India. This is also substantiated by the fact that Ce which is also an important component of automobile catalytic converters (Helmers 1996) shows a very weak loading (0.11) for factor 2 and also very low correlation with Zr and Hf which are also components of autocatalysts. The higher mean levels of these metals obtained for the present work in comparison to those obtained for road dusts from traffic areas of Hyderabad (Mathur et al. 2011) also support this inference. The accumulation of Zr, Hf, Y, and Pb which strongly correlate with factor 2 of PCA (Table 7) can thus be traced back mainly to industrial processes and less significantly to automobile emissions.

Vanadium exhibits a strong correlation with Co, Ce, and Y (Table 6) and is also grouped along with these metals under factor 3 of PCA. Ce and V are fuel additives and along with Co are released due to petroleum and vegetation burning and domestic combustion processes. This inference is in consistence with that of Rodriguez et al. (2004). V, Co, Ce, and Y are markers of industrial emissions as they are used in the manufacture of glass, ceramics, and electronics and in dye making, chemical, and metallurgical processes. They are also employed in different vehicle parts like V and Co that are parts of steel studs of tyres (Edwards et al. 1995; Helmers 1996; Zereini et al. 2001) which implies their release partly due to automobile emissions and wear and tear of automobile parts. V is also released due to abrasion of asphalt. Co, Y, and Zr may have also originated from the parent rocks and are thus considered to have the influence of natural sources (Fergusson 1990; Govil et al. 2008; Wei et al. 2010). Thus,

associations of the studied heavy metals with different products and processes and inter-element correlations (Table 6) and respective factor loadings (Table 7) imply to their origin from multiple sources which could be mainly linked to vehicular and industrial activity and in part to domestic and geogenic/lithogenic sources. I_{geo} , EF and Cⁱ_f results (Figs. 2, 3 and Table 5) also indicate considerable enrichment of these metals in the road dusts and contamination of the study area. Other studies also reported the combined pollution of soils and road dusts by multi-heavy metals (Govil et al. 2008; Krishna and Govil 2008; Wei et al. 2010; Shi et al. 2013).

Conclusion

Excessive levels of heavy metals in the environment severely disturb the natural geochemical cycling of the ecosystem. People in residential areas surrounded by industries are to a large extent exposed to heavy metal-laden polluted road dust which is detrimental to their health. In view of this, the present study was taken up to assess the extent of heavy metal contamination of road dusts from industrial areas of Hyderabad and to delineate their sources. The results obtained reveal that the studied road dusts have elevated concentrations of Cu, Zn, Cr, Co, Pb, Ni, V, Zr, Ce, Y, and Hf. These heavy metals are significantly enriched in comparison to their crustal abundances which demonstrate an intensive impact of various anthropogenic sources on their accumulation. Wide variations of the metal concentrations reflected by their maximum and minimum values also substantiate this finding. Similar trends of mean metal levels obtained applying the three pollution indices (I_{geo} , EF, and C_{f}^{i}) indicate the relative magnitude of contamination of each toxic metal. On the basis of I_{geo} , the road dusts are categorized as practically uncontaminated to moderately contaminated with respect to all metals except Pb. Mean EFs of the studied metals classify the dusts to be moderately to significantly enriched while their mean $C_{\rm f}^{\rm i}$ implies moderate to considerable contamination. Highest mean levels displayed by Pb for the three pollution indices suggest very high level of contamination of this metal in the study area. The results of correlation analysis and PCA also provide the same information on heavy metal accumulation confirming a strong anthropogenic influence from multiple sources, attributable mainly to industrial and vehicular activity, besides they being partly contributed by domestic and natural sources. Release of untreated industrial effluents produced by processing plants and manufacturing of goods, combustion of fuels and emissions from smoke stacks, random dumping of solid waste as well as vehicular exhaust emissions and corrosion/wear and tear of vehicle components appear to have contributed significantly to the accumulation of heavy metals in the road dusts. As there are no designated areas for different types of activities especially due to intermingling of residential settlements and industrial units, emissions from different sources may have carried different metals. The results of this study imply the need for periodic investigation of the study area to understand the spatial distribution and pathways of heavy metals and the relative significance of different emission sources to assess their ecological implications and human health risks. The results can serve as a useful guide for the management to plan effective strategies for pollution control like implementing improved treatment techniques for solid waste and industrial effluents, assigning specific areas/drains for waste disposal, bioremediation, phytoremediation, and instituting strict laws for vehicle fitness and operation. It is suggested that government should establish monitoring stations and launch awareness programs for the public about the health risks associated with the heavy metal pollutants.

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