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Unveiling sources, contamination, and eco-human health implications of potentially toxic metals from urban road dust

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To investigate the pollution characteristics, ecological and health risks assessment, and source apportionment of potentially toxic metal(loid)s (PTMs) in urban road dust, 140 dust samples collected from eight renowned roads of city Lahore, Pakistan. The geo-accumulation index (I_{geo}) and enrichment factor (EF) were used for pollution characteristics, modified ecological risk index used for ecological risk assessment, the USEPA models used for health risk assessment, and multivariate statistical analyses were used for source apportionment of PTMs. The ranges of average concentrations (mgkg^{-1}) in road dust Cd, Hg, Mo, and Pb were 1.91 (CR) to 3.35 (BR), 11.7 (JR) to 29.3 (MuR), 452 (JR) to 1115 (MuR), and 36.9 (MaR) to 110 (BR), respectively, which were several times higher than reference values. The Cd was moderate to highly polluted in road dust with highest I_{geo}=2.58 from MuR and EF = 26.9 from CR. Whereas Hg and Mo were categorized as extremely polluted PTMs with mean I_{geo} and EF values beyond the uppermost level (class 5). The road dust collected from mall road (MaR) was polluted with high levels of most of the PTMs among other roads. The adjacent areas of roads were at extreme ecological risks due to Cd, Hg, and Mo pollution. The employed statistical methods proved that PTMs pollution was induced severely by industrial, exhaustive, and non-exhaustive vehicular emissions in road dust. The Hg pollution in road dust was causing potential non-carcinogenic risks in children with HQing and HI higher than 1. No carcinogenic risk was found for both adults and children. The study helps to create awareness about PTMs' pollution and associated health concerns among public.

Keywords Road dust, Urban pollution, Toxic metals, Pollution characterization, Health risk assessment

Rapid urban expansion and industrial growth stand as the chief instigators of potentially toxic metal(loid)s (PTMs) along with other potential contaminants in the environment of metropolitan areas[1–3]. Whereas these PTMs are ubiquitous in natural matrices and anthropogenic activities like transportation, mining, industrial, and agricultural activities contribute in disturbance to the ecological communities as well as to an escalate PTMs levels in different environmental compartments like soil, air, dust, sediments, and water[4–6]. Due to their less degradable, high bioaccumulative and toxic nature, PTMs persist into environment for long time, potentially leading to irreversible harmful effects on ecosystems and human health. The possibility to intact PTMs with humans can be imperative with the hand to mouth activities, interaction with agricultural and aquaculture products, cause serious cancerous and non-cancerous hazards to human health[7]. The importance of PTMs into environment and exposure to humans has been explained previously with their toxicity effects on human organs, endocrine and immune systems, vascular and skin diseases, etc[8,9]. Vigilant monitoring of PTMs pollution in urban areas is imperative, offering crucial insights into the extent of their contamination and toxicity associated risks to both ecosystems and human health[10,11]. In this context, urban road dust emerges as a promising

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environmental compartment for PTMs pollution assessment, owing to its cost-effectiveness, ease of execution, and swift sample collection process[6,12].

Roads of especially urban areas are crucial to study for environmental sciences with higher chances of particulate suspensions and variety of PTMs sources which can easily be transported via drift of fast-moving vehicles and deposited on the road banks. Road dust has proved one of the eminent environmental media for PTMs pollution assessment and associated health risks in the recent past[13–16]. The sources of PTMs in the road dust from global metropolises are widely distributed and identified as natural crustal matrices, coal combustion, industrial processes, and majorly the exhaustive and non-exhaustive emission from automobiles[16]. As high-temperature fuel combustion processes of biomasses lead to the release of significant amounts of PTMs in the environment[17]. The non-exhaustive emissions of automobiles include remains of tires due to frictions on the roads, corrosion and rusting materials inclusions into road dust by abrasion of vehicles, which were vital contributors of PTMs in the road dust[18].

Various pollution assessment methods, such as the integrated pollution index (IPI), geo-accumulation Index (I_{geo}), potential ecological risk index (PERI), and enrichment factor (EF), have been extensively employed to gauge the threats posed by PTMs based on their concentration, toxicity, and the combined effects[3,19]. These techniques provide diverse perspectives for analyzing contamination and associated ecological risks[20]. Nevertheless, limitations exist for NIPI and RI; NIPI overlooks significant variations in hazardous response variables among heavy metals, and RI may yield inflated results when assessing samples containing multiple heavy metals[21]. Consequently, there is a demand for a novel risk assessment approach that integrates toxic response characteristics while mitigating the influence of heavy metal toxicity[22]. Consequently, in present study we modified already existing IPI and ERI methods to integrate the toxic response effects of the PTMs, as previously employed by[20].

In addition to other challenges, rapid urbanization and industrialization are crucial challenges for a big city like Lahore with elevated anthropogenic input to urban environmental pollution[23]. Consequences of urbanization lead to the unceasing buildup of irregular infrastructure, clustering of small industries, and rush of vehicles on the roads in the metropolitan areas. Lahore is the hotspot among the most populated cities of Pakistan and carries factors mentioned above, making it more challenging to address the environmental issue of PTMs pollution[3]. Additionally, the lack of scientific data, especially on PTMs pollution from road dust, makes the present study more commendable. While elevated concentrations of PTMs have been widely reported in road dust worldwide, only a few studies have been published on road dust PTMs pollution, particularly in Pakistan, with the main focus on large areas per unit sample[23,24]. For instance, Qadeer et al. (2020)[23]found that PTMs pollution in road dust of Lahore and Faisalabad is a severe environmental concern with elevated concentrations of Pb and Zn. Another study on dust-bounded PTMs in various areas of Lahore concluded that dense human population and associated anthropogenic activities are major factors contributing to PTMs contamination in the environment[25]. Whereas the present study focuses on road dust sampled from eminent roads metropolitan areas of Lahore. The study is pioneering to demonstrate the data on PTMs pollution levels and their potential risks to public health and the environment from road dust collected from eight eminent roads of Lahore city. The primary objectives of the present study are as follows; (a) to examine the individual distribution of PTMs concentrations in dust collected from eight prominent roads within Lahore city; (b) to assess the varying levels of PTMs pollution and their potential sources across selected roads using different pollution indexes and statistical analyses; and (c) to evaluate the potential ecological and public health risks associated with PTMs in urban road dust of Lahore, Pakistan.

Materials and methods

Study area

Lahore, located at 31.5204° N, 74.3587° E, and 215 m above sea level, is one of the largest metropolitan cities and the capital of prime Province (Punjab) of Pakistan. A city with enormous cultural and historical heritage with a briskly growing population of more than 11 million, having around 365 km web of roads, which bear the load and friction of more than a million vehicles. The unprecedented pace of urbanization has extended its area up to 1772 Sq. km[26,27]. The climate is hot and humid and comes under the semi-arid region with less rainfall. A dense population, shedload of vehicles on roads, and numerous patches of small and big industrial clusters are always putting this city on the top-ranked polluted hotspot on the globe. Eight prominent roads of Lahore city, including Ravi Road (RR), Band Road (BR), Mall Road (MaR), Jail Road (JR), Multan Road (MuR), Wahdat Road (WR), Ferozepur Road (FR), and Canal Bank Road (CR) were selected for dust sampling. The location points with respect to selected roads for dust sampling on the map are depicted in Fig. 1. These roads experience high vehicle density around the clock, with light vehicles primarily operating during the daytime and heavy vehicles predominantly active at night. This primarily contributed to the input of PTMs in road dust through both exhaustive and non-exhaustive vehicular emissions while having minimal impact from other land use characteristics in the surrounding area. However, the local land use characteristics of these roads that correspond to several activities are comprehended in Table 1.

Sampling strategy

Plastic brushes and dustpans were used to collect the samples by gently sweeping the terrestrial/visible dust from a specific area of 1 $m^2$of the road surface[28,29]. A representative composite sample of road dust was made by mixing three samples taken from different points of each sampling location. The pebbles, stones, and other unwanted debris were removed manually before transferring the collected samples to the zip-locked polythene bags and carefully transported to the laboratory. All the equipment was rinsed three times with distilled water and then dried with the help of clean cotton. The cleaning procedure was repeated after taking each sample to avoid any unnecessary contamination. A total of 140 dust composite samples were collected from the banks of

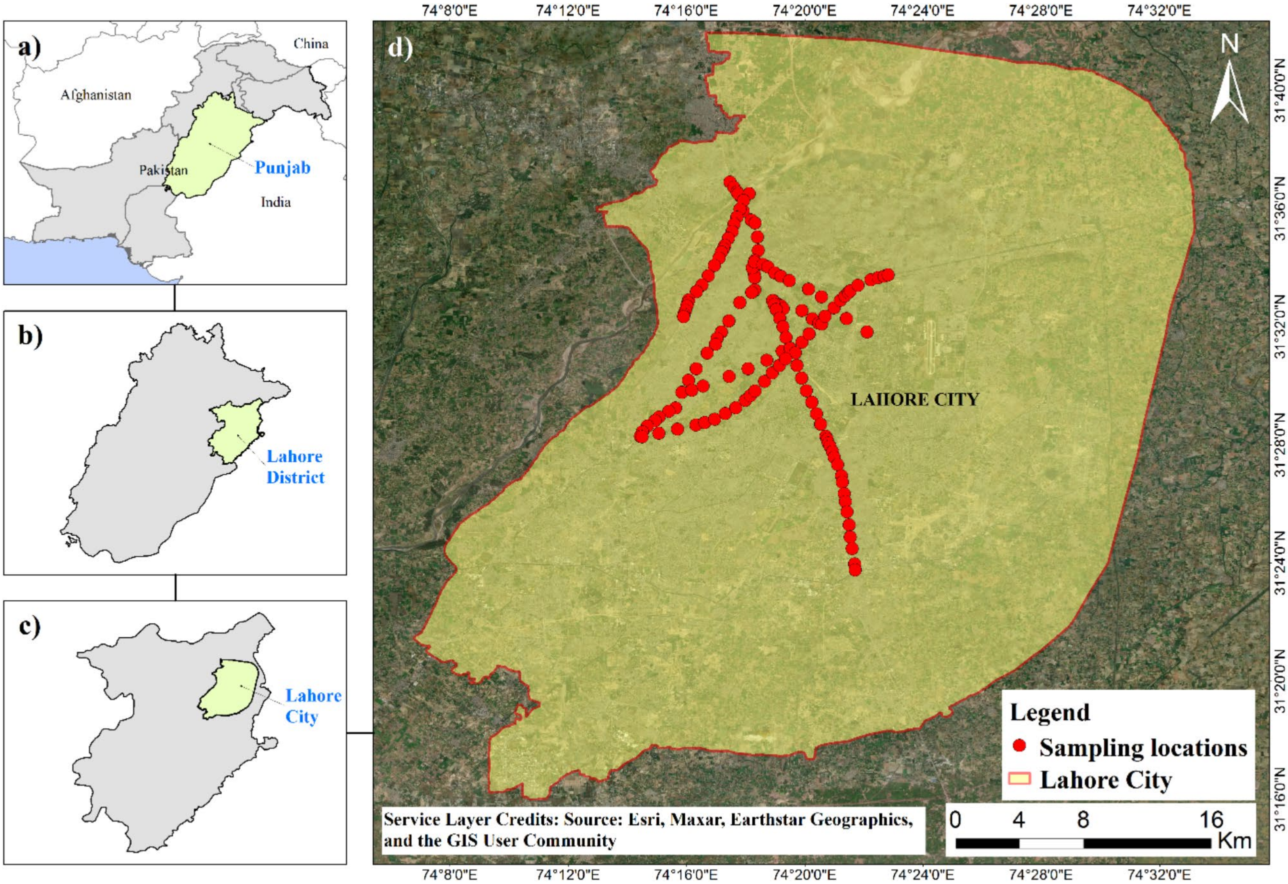

Fig. 1. Study area of Lahore, Pakistan with sampling locations of dust collection from prominent roads of the city. The map was created using ArcMap 10.8, incorporating satellite imagery (Basemap; Esri, Maxar, Earthstar Geographics, and the GIS User Community), while data on Pakistan's administrative boundaries was sourced from https://data.humdata.org/.

Roads/Land-use	Residential	Commercial	Industrial	Institutional	Vehicle density	VRS
Ravi Road	-	++	+	-	++	+
Band Road	+	++	++	-	++	++
Mall Road	-	++	-	++	++	+
Jail Road	++	++	-	+	++	+
Multan Road	++	++	+	++	++	++
Wahdat Road	++	+	-	++	++	+
Ferozepur Road	++	++	+	++	++	++
Canal Road	++	+	-	++	++	-

Table 1. Description of local land use characteristics for prominent roads of Lahore. VRS = vehicular repairing shops; - = no to less extent, + = moderate extent, ++ = high extent.

the selected roads. The general information about sampling collection and roads, i.e., starting and ending points, length (km) of roads considered for sampling, and number of samples with respect to each selected road, are given in Table S1. Whereas the coordinates of sampling locations were also noted and given in Table S2.

Chemical and instrumental analysis

The collected samples were oven dried at 65˚C for a day prior to sieve through 100 µm nylon mesh. Finer dust particles have high susceptibility to carry toxic elemental fractions, as more finer the particulate could easily access to the living beings to cause hazards[30]. The standard acid digestion method has frequently been utilized to chemically break down the mineral and organic fractions in soil and dust samples, allowing for the extraction and analysis of residual PTMs concentrations[29,31]. Therefore, aqua regia solution was prepared with the help of 1:3 of two strong acids, i.e., HNO_3 and HCl. Each road dust sample was weighed by 0.2 g and transferred into the

50 ml volumetric flask. In each weighed sample, 15 ml of aqua regia solution was added and placed for overnight in the fume hood. Next day, all the samples were placed on a hotplate at 210˚C for complete digestion of the dust fractions. Upon near-to-dry state of the digested samples, 5 ml of deionized (DI) water was added and heated at 100% until dryness. The DI water was then added to make 20 ml volume of each sample. All the samples were filtered and stored in the refrigerator for instrumental analysis. Twelve toxic metals (i.e., As, Cd, Co, Cr, Cu, Fe, Hg, Mo, Ni, Pb, Sb, and Zn) were selected to analyze from the prepared samples with the help of Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES, 2100DV, Perkin Elmer Optima).

Quality assurance

Certified reference material (NIST SRM 1633) was used to ensure the quality control and precision for this study. The recovery range of elements in the certified reference material was within the acceptable range of 86.7% and 103.8%. Blank samples (without road dust) and some of duplicates were prepared and used after every set of ten samples to ensure the precision and stability of the data. No metals were detected in the blank samples and duplicate samples indicated the acceptable range in accordance with the quality assurance criteria.

Pollution characterization

In order to determine the degree of PTMs pollution of road dust of Lahore, two well established methods i.e., geo-accumulation index (I_{geo}) and enrichment factor (EF) were employed in present study. Briefly, I_{geo}method helps to determine the degree of PTMs pollution with respect to background or reference concentrations of respective PTMs[32]. However, EF can explain the contamination level of PTMs with respect to influence of anthropogenic and/or natural resources[33,34]. In I_{geo}calculations, a constant value is involved to overcome the lithogenic influence and/or fluctuations. Whereas, a reference element is involved in the calculations of EF, that element considered to be geochemically stable element in the environment and either of Al, Fe, Sc, and Ti[35]. The reference background concentrations in present study for the calculation of $I_{geo}$and EF were considered the values of selected PTMs for shale in upper continental crust (UCC) given in[36]. Since road dust contains natural soil particles yet influenced by anthropogenic activities and can conceivable deposited to bare surfaces at the roadside areas, the values of natural shale of UCC can be used as a background reference to estimate the anthropogenic input of PTMs[25,37–39]. In present study, Fe was used as reference element to calculate EF of PTMs in road dust. The formulae for both I_{geo} and EF are given as;

$$Igeo = \log_2 \left(C_i/kB_i\right) \tag{1}$$

Where, C_i is the concentration of ith PTM, k is a constant value equals to 1.5, and B_i is the background reference concentration of respective PTM.

$$EF = \frac{\left(C_i/C_{Fe}\right)_s}{\left(C_i/C_{Fe}\right)_b} \tag{2}$$

Where, the $(C_i/C_{Fe})_s$ is the ratio of concentration of the ith PTM to concentration of Fe in the sample, and the $(C_i/C_{Fe})_b$ shows the ratio of concentration of the ith PTM to concentration of Fe of the considered background reference values.

Modified ecological risk index

Pollution index (PI) can be used to assess the degree of pollution of PTMs, which based on the background or reference values of PTMs. Likewise, ecological risk index also used widely for PTMs. In present study, modified integrated pollution index (MIPI) and modified integrated risk index (MIRI) were used to assess the ecological risk of PTMs from road dust of Lahore. Below given formulae can be used to calculate PI, MIPI, Er_i, and MIRI;

$$PI = C_i/B_i \tag{3}$$

$$MIPI = \sqrt{\left(PI_{max}^2 + PI_{avg}^2\right)/2} \tag{4}$$

$$Er_i = Tr_i \times \ PI \tag{5}$$

$$MIRI = \sqrt{\left(Eri_{max}^2 + Eri_{avg}^2/2\right.} \tag{6}$$

Where, C_i is the concentration of ith PTM; B_i is the background value of ith PTM; PI_{max} and PI_{avg} are the maximum and average values of calculated PI values; Er_i is the ecological risk factor of ith PTM; Tr_i is the toxic response factor of ith PTM; and Eri_{max} and Eri_{avg} are the maximum and mean of calculated Er_i values. The toxic response factor used to calculate the Er_i for studied PTMs i.e., As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, and Zn were 10, 30, 5, 2, 5, 40, 15, 5, 5, 7, and 1, respectively. However, MIPI and MIRI were calculated by taking maximum and mean values for individual PTM separately as well as for cumulative of all PTMs.

The description and classification of pollution levels and respective values ranges of I_{geo}, EF, MIPI, and MIRI are given in Table S3.

Multivariate statistical analyses

In this study, Pearson's correlation and hierarchical cluster analyses were employed to delve into the interrelationships among studied toxic metals. This approach was crucial as it allows to uncover the intricate connections between PTMs, thereby providing valuable insights into their source of origins. Additionally, Principal Component Analysis (PCA) was also used to further explore the potential sources of PTMs present in the road dust[6]. PCA, a receptor model grounded in the principle of mass conservation, has widely been recognized for its effectiveness in investigating potential source attributions of particulate matter[16]. However, this statistical approach of combining three different analytical methods to find inter-relationships among PTMs in road dust proved reliable to clarify and determination of potential sources[19]. The KMO factor to check data adequacy to perform PCA was 0.77, indicating average to good adequacy. The normality (Kolmogorov-Smirnov test) and homogeneity (Levene's test) tests of the data were conducted using SPSS (version 27) and concluded in Table S4&S5. However, the study area map was developed using ArcMap 10.8 to integrate satellite imagery (Basemap: Esri, Maxar, Earthstar Geographics, and the GIS User Community), and the data on the administrative boundaries of Pakistan was obtained from https://data.humdata.org/.

Health risk assessment

The US-EPA models were employed to assess the cancerous and non-cancerous health risks in children and adults caused by PTMs from road dust of Lahore, Pakistan. Humans can be exposed to road dust associated PTMs through three routes, namely ingestion, inhalation, and dermal contact[40–42]. Road dust carrying toxic metals has a fair bit of chance to suspend in the air as vehicles hovering around and access humans via those aforesaid routes. In present study, non-carcinogenic risk was assessed for As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Sb, and Zn. However, for carcinogenic risk assessment, only As, Cd, Co, Cr(VI), Ni, and Pb were taken into account for children and adults. The hexavalent Chromium (Cr^{6+}) was considered as carcinogenic and its concentrations was obtained by dividing the total elemental concentration of Cr by 7, as both species of Cr i.e., Cr(III) and Cr(VI) has been assumed to be at the ratio of 1:6 in the environment[43]. The important terms for health risk assessment can include; average daily exposure dose (ADD), hazard quotient (HQ), hazard index (HI), and cancer risk (CR). Whereas, their formulae for calculations are given below;

$$ADD_{ing} = C \times BA \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{7}$$

$$ADD_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \tag{8}$$

$$ADD_{der} = C \times \frac{SL \times SA \times ABF \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{9}$$

Where, ADDing, ADDinh, and ADDder represent the average daily exposure dose via ingestion, inhalation, and dermal contact, respectively. The definitions and values of several factors used in aforementioned equations are given in Table S6[44].

The equations used in the calculation of HQ, HI, and CR for PTMs are as follows:

$$HQ = ADD/RfD \tag{10}$$

$$HI = \sum HQ \tag{11}$$

$$CR = ADD \times SF \tag{12}$$

Where, RfD is the homologous reference dose and SF is the homologous slope factor. In this study, the values of RfD and SF for selected PTMs were given in Table S7[45–47].

Results and discussion

Pollution characterization of road dust

PTMs concentration

The concentrations of 11 selected PTMs in the dust collected from eminent roads of Lahore were estimated by ICP-OES and their statistics tabulated in Table 2. The international standard limits of PTMs set by neighboring countries (i.e., China and India), WHO, and background values were also given. The comparison among various reported studies on PTMs concentrations in road dust samples is given in Table 3. Among other PTMs, the mean concentration of As was comparatively found within the permissible limits set by international guidelines in all selected roads dust. The average contents of Cr were measured in the dust of MaR (149 $mgkg^{-1}$), JR (222 $mgkg^{-1}$), and MuR (101 $mgkg^{-1}$), which were higher than corresponding background and WHO standard guidelines. The mean concentration of Cu was higher in RR (46.6 $mgkg^{-1}$), BR (70.7 $mgkg^{-1}$), MuR (57.7 $mgkg^{-1}$), and FR (45.9 $mgkg^{-1}$) than corresponding background values. however, Zn concentration was also exceeding the corresponding background values from the dust of RR (141 $mgkg^{-1}$), BR (228 $mgkg^{-1}$), MuR (114 $mgkg^{-1}$), and FR (148 $mgkg^{-1}$). However, concentrations of Cd, Hg, and Mo were observed predominantly higher than other PTMs in all selected roads of Lahore. The mean concentrations of Cd and Hg were found; 8 and 54 times higher in RR dust; 11 and 65 times higher in BR dust; 7 and 45 times higher in MR dust; 9 and 29 times higher in JR dust; 8 and 73 times higher in MuR dust; 7 and 63 times higher in WR dust; 8 and 49 times higher in FR dust; 6 and 51 times higher in CR dust, respectively, as compared to corresponding background values. The concentration of Mo was measured; 23 times higher in RR dust, 18 times higher in BR dust, 15 times higher in MaR dust, 11 times higher in JR dust, 28 times higher in MuR dust, 14 times higher in WR dust, 15 times higher in FR dust, and 12 times higher in CR dust as compared to WHO recommended standard value. The prominent roads among selected for dust sampling with exceeding PTMs concentrations were RR, BR, MuR, and FR. On

Concentration	As	Cd	Co	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Zn
Ravi Road (RR) ($n=13$)											
Min	9.96	1.58	6.92	36.3	16.1	14.6	380	12.3	26.5	2.13	67.2
Max	14.1	3.81	9.41	95.1	79.1	27.1	1332	30.4	161	4.88	329
Mean	12.2	2.29	8.31	54.5	46.6	21.6	932	20.9	78.6	3.57	141
Band Road (BR) ($n=21$)											
Min	0.93	0.81	6.29	19.8	12.4	4.70	242	12.1	1.50	0.03	24.1
Max	18.7	12.3	31.1	380	319	47.8	1488	115	684	19.4	897
Mean	10.1	3.35	12.6	89.5	70.7	26.1	733	33.3	110	5.49	228
Mall Road (MaR) ($n=13$)											
Min	3.80	1.92	7.38	68.4	15.2	0.78	245	11.3	22.7	1.22	7.68
Max	14.4	2.62	10.2	330	42.2	32.8	1094	23.9	59.1	8.02	102
Mean	9.84	2.19	8.99	149	28.4	18.3	586	16.7	36.9	4.33	55.9
Jail Road (JR) ($n=7$)											
Min	9.82	1.83	7.43	63.5	20.1	1.13	230	13.2	27.6	1.73	48.9
Max	15.1	3.37	9.83	535	66.8	22.3	897	26.4	74.7	12.6	118
Mean	11.9	2.60	8.91	222	42.1	11.7	452	20.5	51.7	6.40	87.2
Multan Road (MuR) ($n=18$)											
Min	6.52	1.70	7.32	36.1	18.7	8.23	154	12.4	28.1	2.07	52.7
Max	14.9	3.62	13.9	270	124	52.0	1586	29.4	132	8.30	213
Mean	11.9	2.43	8.92	101	57.7	29.3	1115	20.6	57.4	4.06	114
Wahdat Road (WR) ($n=8$)											
Min	3.17	1.95	7.65	48.4	22.9	8.23	58.7	17.0	27.1	1.13	74.7
Max	13.2	2.48	9.75	136	40.8	39.6	1438	20.7	54.6	4.88	114
Mean	9.45	2.16	8.66	86.0	30.7	25.4	565	18.2	41.1	3.02	85.4
Ferozepur Road (FR) ($n=31$)											
Min	2.87	1.43	6.06	33.9	17.4	4.34	129	13.4	37.6	1.20	77.6
Max	19.8	4.49	15.8	91.1	87.2	33.0	1541	43.0	137	6.55	352
Mean	10.6	2.27	9.35	56.7	45.9	19.6	612	21.8	66.5	3.21	148
Canal Road (CR) ($n=29$)											
Min	2.88	1.47	6.26	31.6	14.6	2.52	51.2	13.6	17.3	0.15	41.7
Max	19.3	2.78	11.4	59.7	70.3	40.6	1174	33.6	143	4.10	172
Mean	12.7	1.91	7.79	43.1	29.9	20.3	490	18.4	46.0	1.81	80.4
Permissible Standards											
China [a]	30.0	0.60	13.5	200	100	NA	NA	50.0	300	NA	200
WHO [b]	20.0	3.00	50.0	100	100	NA	40.0	50.0	100	NA	300
India [c]	NA	3–6	NA	NA	135	NA	NA	75.0	250	NA	300
BG [d]	13.0	0.30	19.0	90.0	45.0	0.40	2.60	68.0	20.0	1.50	95.0

Table 2. The concentration of toxic metals in road dust of Lahore. a = [48], b = [49], c = [50], d = [51].

the contrary, Cd, Hg, Mo, Sb, and Pb were prominently high in concentrations among selected PTMs from road dust of Lahore.

Geo-accumulation index

The comprehensive depiction of calculated I_{geo} for selected PTMs were given in Fig. 2. The mean I_{geo} values of PTMs like As, Co, Cr, Cu, Ni and Zn were found to be less than 0 showing no accumulation in dust of all selected roads of Lahore. According to calculated I_{geo} values, Pb and Sb were found in class 1 showing low accumulation in almost all the selected roads. Whilst some of the dust samples collected from JR, MuR, and WR with high I_{geo} values which slightly made the mean values of 1.01, 1.29, and 1.15, respectively, into class 2 showing moderate accumulation of Pb. The Cd fall in the class of moderately to highly accumulated PTM in all the selected roads with mean I_{geo} values of 2.43, 2.36, 2.26, 2.12, 2.58, 1.96, 2.07, and 2.45 for RR, BR, MaR, JR, MuR, WR, FR, and CR, respectively. For most of the roads, the Hg was laid in the range of class 5 with high to extreme accumulation level. For Hg, the calculated mean I_{geo} values were 4.05, 4.75, and 4.79 for RR, FR, and WR, respectively, indicating high to extreme accumulation, whereas, with mean I_{geo} values 5.06, 5.12, 5.22, 5.27, and 5.49 for MuR, BR, JR, CR, and MaR showing extreme accumulation of Hg. At last, the Mo showed extreme accumulation in all road dust samples with I_{geo} more than 6. However, the descending order of selected roads with respect to mean I_{geo} of Mo was MaR > JR > MuR > CR > RR > BR > FR > WR.

Study area	PTMs concentration in road dust ($mgkg^{-1}$)										
	As	Cd	Co	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Zn
Present study[a]	11.1	2.40	9.19	100	44.1	21.5	686	21.3	61.1	3.99	117
Sharjah, UAE[b]	0.97	1.25	-	460	150	-	-	856	68.3	-	392
Datong, China[c]	12.3	0.47	11.8	109	32.1	0.09	-	30.3	31.6	-	172
Zunyi, China[d]	-	-	16.2	127	134	-	-	43.2	86.8	-	507
Islamabad, Pakistan[e]	-	5.0	-	-	52	-	-	23	104	-	116
Lahore, Pakistan[f]	-	1.4	-	-	23.9	-	-	13.9	44.3	-	67.9
Faisalabad, Pakistan[f]	-	1.3	-	-	25.7	-	-	11.4	38	-	64.1
Charsadda, Pakistan[g]	-	5.89	44.8	16.2	119	-	-	52	338	-	908
Jharkhand, India[h]	70.2	1.2	21	167	1146	-	-	87.3	112	-	947
Busan, South Korea[i]	13.3	1.1	8.7	92.2	80.7	0.01	-	15.6	85.3	5.4	430
Huainan, China[j]	12	0.64	2.94	83	57	0.26	-	23	71	-	828
Dhaka, Bangladesh[k]	8.09	11.6	-	144	49.7	-	-	37.1	18.9	-	239
Jeddah, Saudi Arabia[l]	21.6	7.46	11.7	65.4	139	-	-	11.7	141	-	488

Table 3. Comparison of PTMs concentrations in road dust reported in recent literature nearby countries. a = average concentration of PTMs in dust from all selected roads. b =[58], c =[59], d =[60], e =[24], f =[23], g =[61], h =[38], i =[62], j =[63], k =[64], l =[65].

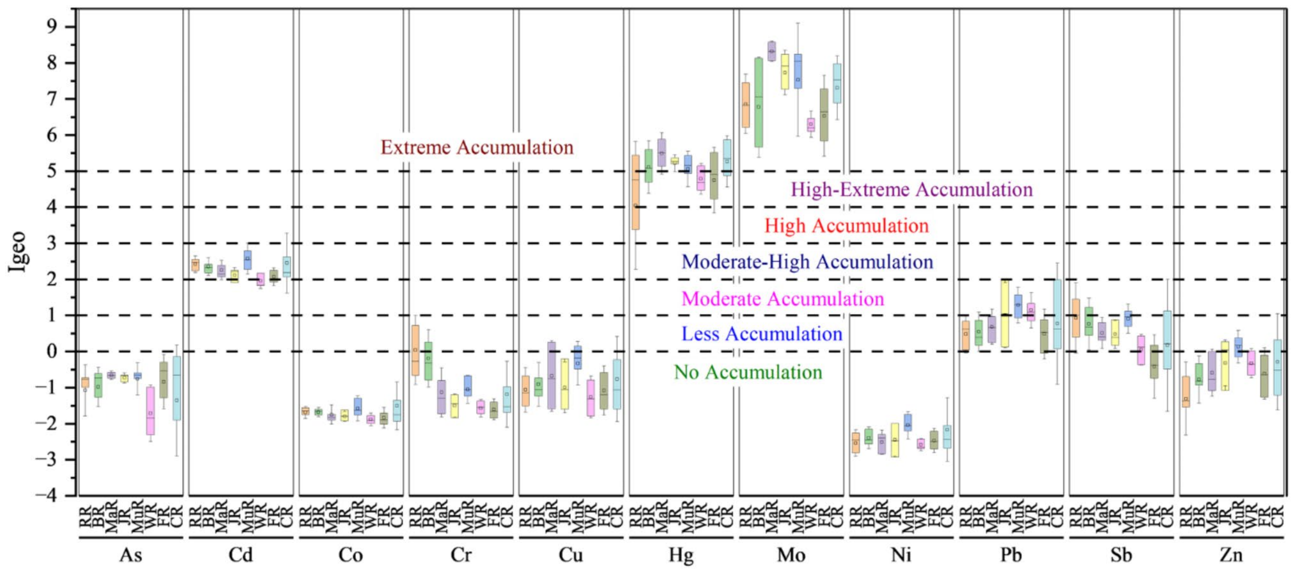

Fig. 2. Depiction of geo-accumulation index (I_{geo}) calculations for PTMs in road dust and their corresponding classification at different levels of accumulation with respect to selected roads of Lahore, Pakistan.

Enrichment factor
The calculated EF values and corresponding enrichment levels of PTMs from road dust samples of Lahore were showed in Fig. 3. The mean EF values for Co and Ni were below 2 for all the selected roads, indicating their minimum enrichment in the road dust. In cases of As, Cr, Cu, and Zn, with mean EF values in between 2 and 5, indicating their moderate enrichment to some extent in all selected roads of Lahore. Moreover, Pb and Sb showed significant enrichment in dust collected from all the selected roads with mean EF values in the range of 5–20. The mean EF values of Sb in dust collected from FR and CR were 4.25 and 5.60, respectively, showing moderated enrichment. Moreover, the most prominent PTMs were Hg and Mo, which has extreme levels of enrichment into road dust and much higher mean EF values for all selected roads of Lahore.

Modified integrated pollution index (MIPI)
Modified integrated pollution index was employed to estimate the contamination of PTMs in road dust in accordance with their corresponding background values. As given in Fig. 4a, the mean PI values of As from all selected roads of Lahore were lies very close to 1 which indicated that As was in the warning limit of pollution. In contrast, the MIPI values for As had slightly been crossed 1, which indicates less degree of pollution in dust of all roads. The calculated IPI values of Cd, Hg, and Mo were away beyond the standard limit of extremely polluted range for dusts collected from all the roads of Lahore. Among all PTMs, only Co and Ni were observed to have

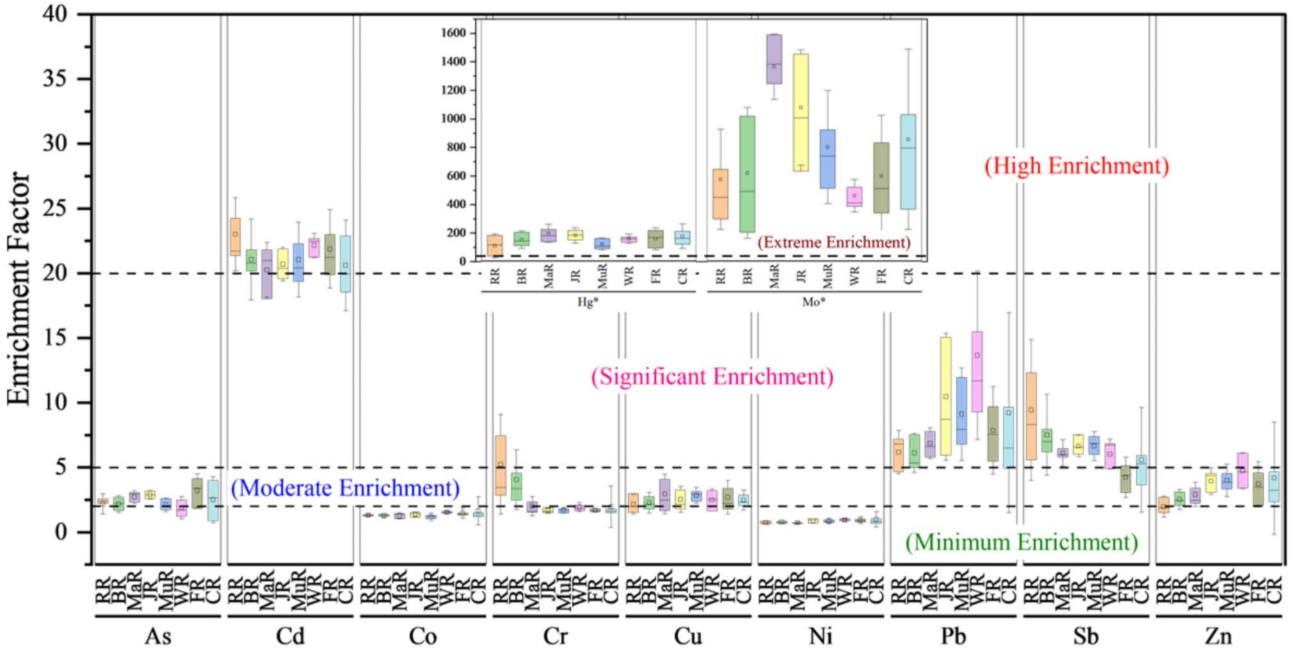

Fig. 3. Depiction of enrichment factor calculations for PTMs in road dust and their corresponding classification at different levels of enrichment with respect to selected roads of Lahore, Pakistan.

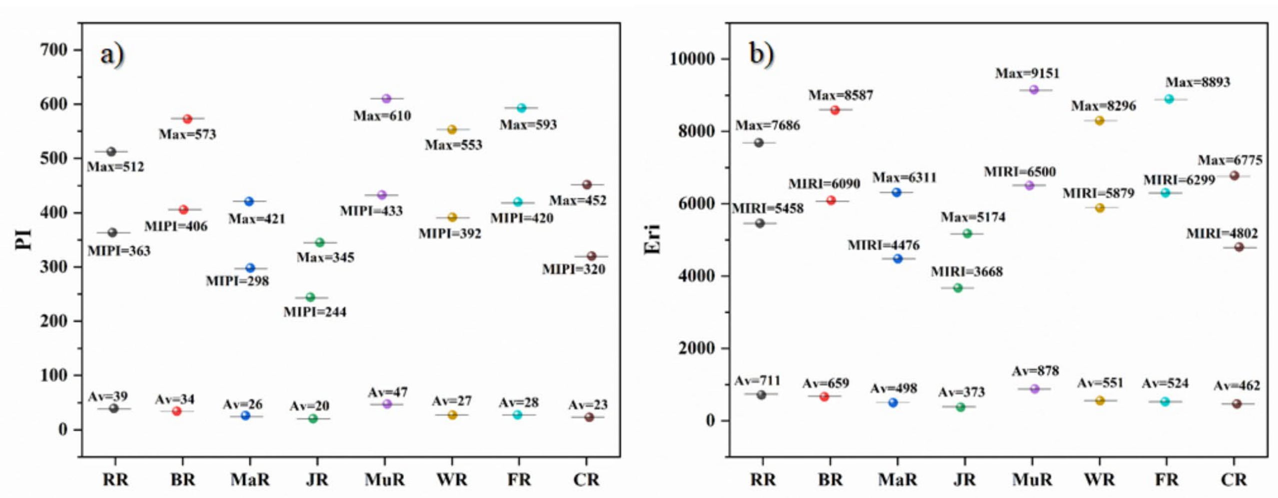

Fig. 4. (a) The maximum (Max) and mean (Av) values of Pollution Index (PI) along with modified integrated pollution index (MIPI) values; (b) The maximum (Max) and mean (Av) values of Ecological Risk Index (Eri) values along with modified integrated risk index (MIRI) values, to cause pollution and potential risks by PTMs from dust with respect to selected roads.

mean PI and MIPI less than 1 in dust collected from all studied roads, except the IPI values from dust of BR i.e., 1.25 and 1.24 for Co and Ni, respectively. Which ultimately the indication of no to less pollution of Co and Ni. The mean IPI values indicated no to less pollution of Cr in RR (0.86), FR (0.84), and CR (0.58); moderate pollution in BR (3.07), MaR (2.84), MuR (2.26), and WR (1.26); and slightly high pollution in JR (4.55) dust. The Cu in road dust was categorized as unpolluted in MaR (0.80) and WR (0.80); less polluted in RR (1.44), JR (1.24), MuR (2.15), FR (1.55), and CR (1.20); and moderately polluted in BR with IPI values of 5.14. The Pb was moderately to highly polluted in MaR (2.46), JR (3.21), WR (2.42), MuR (5.08), FR (5.39), RR (6.34), and CR (5.31) with IPI values, while, extremely polluted in the dust of BR with IPI value (24.5) beyond 6. The IPI values for Sb indicated moderate pollution in RR (2.85), WR (2.71), and CR (2.11); high pollution in MaR (4.29), MuR (4.36), and FR (3.44); and extreme pollution in BR (9.49) and JR (6.66). However, IPI calculations showed Zn have no to less pollution in the dust of MaR (0.86); extreme pollution in BR (6.89); and except those two roads, all other studied roads were moderately polluted with Zn, as IPI values for these roads were lies in between 1 and 3.

Modified integrated risk index (MIRI)

The ecological risk index assists to evaluate the impact that can likely to pose adverse risks on the ecological system. The results of the MIRI are given in Fig. 4b. The values of MIRI were slightly higher than mean values of Eri showing no significant difference. The MIRI for Cd, Hg, and Mo in dust of all selected roads were comparatively higher than other PTMs. The MIRI for Cd showed high ecological risks in BR (314), MuR (308), JR (301), MaR (242), CR (238), and WR (233); as their Eri values laid in between 160 and 320 range. However, Cd was posing extreme ecological risks in Ferozepur Road and Band Road with MIRI values beyond 320, i.e., 356 and 902, respectively. The Eri values for Hg and Mo in all selected roads were larger than 320 showing extreme risks. Moreover, the dust collected from Band Road showed considerable ecological risk due to Pb with MIRI = 123 and moderate risk due to Sb with MIRI = 66. Whereas rest of the PTMs in dust of almost all selected roads showed MIRI values less than 40 showing low ecological risks.

Source identification

The study employed a trio of statistical analyses including Pearson's correlation, cluster analysis (CA), and principal component analysis (PCA) to unearth potential sources of PTMs in the road dust of Lahore. The results from these analyses coincided and illustrated in Fig. 5. Pearson's correlation of PTMs (Fig. 5a) revealed significant ($p > 0.01$) correlations of Cd and Co with most of the other studied PTMs, except As and Mo. Notably, Cd exhibits a strong relationship with Cu (0.86), Ni (0.85), Sb (0.68), and Co (0.61), indicating a shared source of origin. Additionally, Pb and Zn show a robust positive association with each other, as well as with Co, Cd, and Sb. However, As demonstrates a positive correlation with Hg (0.39) and Mo (0.40) at a significant level of 0.01. Furthermore, the results of CA and PCA, depicted in Fig. 5 (b&c), align well with the findings of Pearson's correlation. For instance, As, Hg, and Mo group together in the first cluster in CA and were found prominently in component 2 with an eigenvalue of 1.99 and 18.1% of the variance in PCA. Similarly, Pb, Zn, and Co emerge as the primary PTMs in component 1, with the highest eigenvalue of 5.21, explaining 47.4% of the variance. This

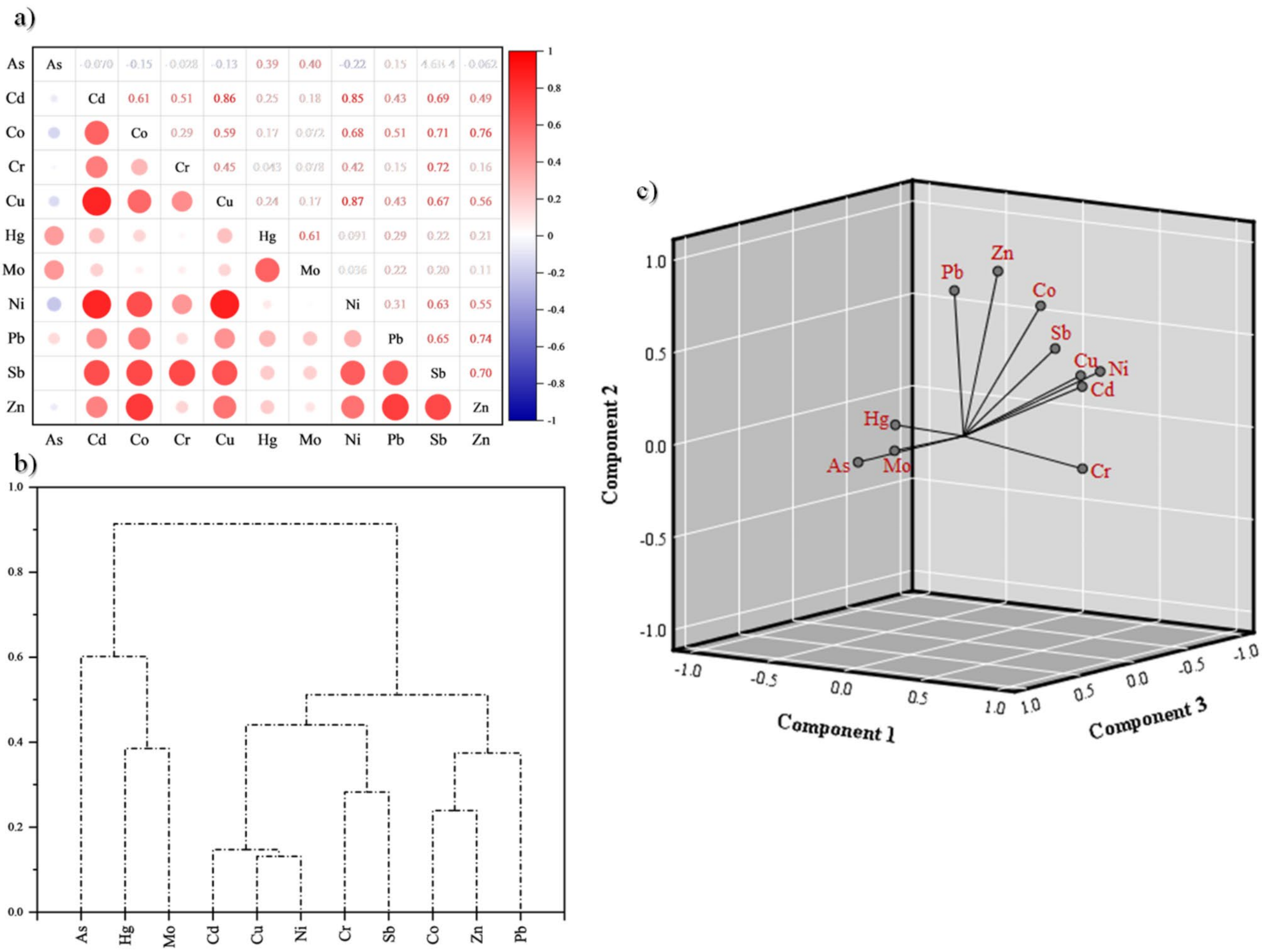

Fig. 5. Source identification of PTMs in road dust of Lahore by (a) Pearson's Correlation plot; (b) dendrogram Cluster Analysis; and (c) Principal Component Analysis.

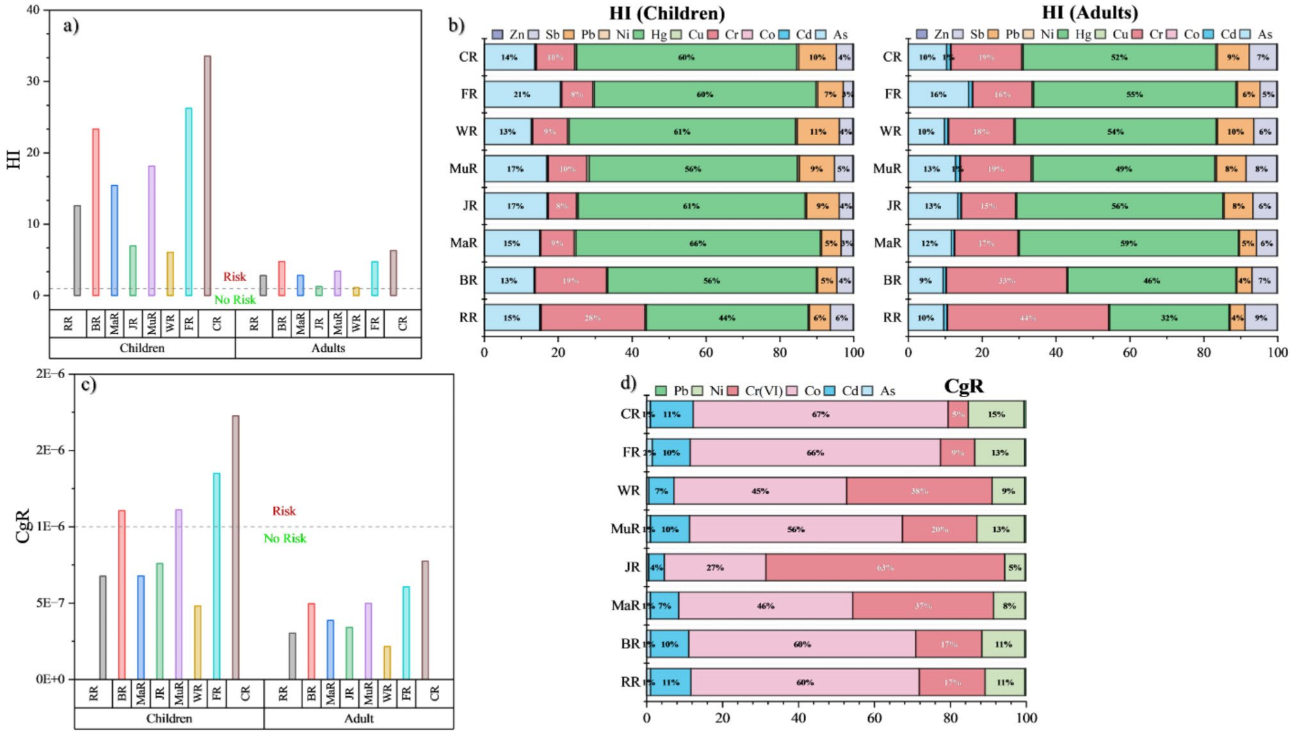

Fig. 6. Bar plots representing hazard index (HI) and carcinogenic risk (CgR) for children and adults posed by overall PTMs from dust with respect to selected roads of Lahore. (a) HI; (b) Percent contributions of PTMs to HI; (c) CgR; (d) Percent contributions of PTMs to CgR.

grouping was further corroborated by CA, which shows these PTMs clustered closely together. Further details of principal components and their respective values for PTMs loadings, eigenvalues, and variance (%) were presented in Table S8, while the KMO factor (0.77) indicated average to good adequacy for PCA. The significant synergy observed among the various studied PTMs in this study can be attributed to identical emission sources.

Health risk assessment

Non-carcinogenic risk

Health risk assessment due to PTMs in road dust of Lahore was estimated using US-EPA models through ingestion, inhalation, and dermal contact pathways for children and adults. Hazard quotient (HQ) and hazard index (HI) were calculated with average daily exposure (ADD) concentration of ten PTMs i.e., As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Sb, and Zn from road dust used for non-carcinogenic risk assessment. The calculated values of ADD and HQ for non-carcinogenic risks of PTMs from road dust of Lahore were depicted in Fig. S1-S4 for children and adults. Whereas the calculation of HI for adults and children through the abovementioned routes with respect to selected roads were well depicted in Fig. 6a. while, the contribution percentage of PTMs to overall HI depicted in Fig. 6b. The HI in all selected roads was found beyond the safer limit for both children and adults, except for JR and WR which showed HI close to 1 for adults (Fig. 6a). However, Hg was the most prominent PTM to contribute more than 50% in HI followed by Cr and As (Fig. 6b). The results for both adults and children were quite identical in terms of PTMs' exposure and causing risks. For instance, in cases of both adults and children, Hg through ingestion pathway and Cr through inhalation and dermal contact pathways were prominent showing higher HQ as compared to other studied PTMs. The Hg was quite lethal to children through ingestion pathway having HQ beyond the safer limit (1.00E + 00). The HQ of Hg calculated most for dust collected from MaR (2.23E + 00) followed by CR (2.04E + 00), BR (1.86E + 00), JR (1.81E + 00), MuR (1.68E + 00), FR (1.50E + 00), WR (1.38E + 00), and RR (1.26E + 00). For adults, the order of HQing for Hg was identical as the children but the value ranges from 3.11E-01 for Mall Road dust to 1.68E-01 for Ravi Road dust, indicating slight non-carcinogenic risks. The rest of the PTMs were also showing HQing close to the permissible limit which means further increment of PTMs concentrations would be threatening for children health. The Cr had HQing values for children of 7.44E-01 in Ravi Road dust and 5.83E-01 in Band Road dust, which were very close to 1. Arsenic (As) also showed its cruciality for non-carcinogenicity in children after Hg and Cr. Cumulatively taking account all the selected roads, the descending order of studied PTMs for HQing of both adults and children was as Hg > Cr > As > Pb > Sb > Zn > Cd > Co. In cases of HQinh and HQder, the calculated values of all the PTMs were distant a bit from the safer limit for both adults and children, indicating no adverse health risks. However, Cr and Co through inhalation pathway; and Cr, Hg, and Sb thorough dermal contact were prominent among other studied PTMs.

Carcinogenic risk

In present study, the PTMs i.e., As, Cd, Co, Cr(VI), Ni, and Pb were selected for carcinogenic risk assessment, which were among the carcinogenic toxic metals indicated by IARC and WHO[52]. The carcinogenic risk was calculated using USEPA recommended method only through inhalation pathway. The overall results of carcinogenic risk (CgR) with respect to roads were depicted in Fig. 6c and the percentage contributions of PTMs to the CgR presented in Fig. 6d. In the case of adults, no carcinogenic risk was found by any of the PTMs in dust collected from any of the selected roads. However, for children, the CgR values were found in the unacceptable range (1E-4 to 1E-6) in dust collected from CR, FR, BR, and MuR. The contribution of Co was observed to be highest in most of the selected roads i.e., 67% in CR, 66% in FR, 60% in BR and RR, and 56% in MuR, followed by Cr(VI), which were highest in JR (63%), WR (38%), and MaR (37%) (Fig. 6d).

Discussion

Pollution and source of PTMs

The present study aimed to investigate the PTMs concentrations, their pollution characteristics, and health risk assessment from dust with respect to eight prominent roads of Lahore, Pakistan. The overall results of PTMs concentrations and calculated pollution indices used in the study coincided with each other, indicating similar kinds of trends. Cumulatively, all the pollution indices (i.e., I_{geo} and EF) indicated extreme pollution of Mo and Hg; moderate to high pollution of Cd; and less pollution of the rest of the PTMs. Unlike other studies, the present study involved distinct elements like Hg and Mo, which were found to be extremely enriched and accumulated by anthropogenic input in the urban road dust (Table 3). The methods used in the study are well documented for the purpose of pollution level descriptions of PTMs from various compartments of environment[24,53–55]. Geo-accumulation index, firstly developed by Muller, (1969)[32], is a well-renowned method of evaluating pollution levels of contaminants like PTMs. Many researchers have been benefitted with accomplishment by utilizing this model, as its calculations involve the comparison with background values of PTMs[9]. This method gives seven reference classes to explain different pollution levels of PTMs (Table S3). The overall pollution status of PTMs was assessed by supporting the information of I_{geo} with the evaluation of enrichment factor calculations. The EF can be served as a prefatory indicator of PTMs enrichment and their causes in the environment[56]. Since EF values above 1.5 indicate the involvement of human interventions in the high concentration of metal, EF values between 0.5 and 1.5 predict that metal pollution is solely caused by lithogenic or geogenic processes[57]. However, EF > 2 strongly favors the accumulation of metal by anthropogenic sources, EF values ranging from 1 to 2 also suggest that the enrichment of metal may be subject to variations in the pedo-geochemical characteristics of soil[56].

Urban areas are known to contain PTMs like Cd, Cr, Cu, Pb, Hg, and Zn, and their elevated concentrations are ultimately the result of anthropogenic activity[27,66]. The results of the present study showed moderated levels of pollution for Cd and extremely higher levels of pollution for Hg and Mo from the road dust of Lahore, Pakistan. The extensive use of rechargeable batteries; plastic and metallic ware of vehicles; and paints and pigments could be the sources of these PTMs like Cd, Pb, Hg, Zn, and Mo in the road dust of Lahore[67]. The usage of Cd in the plating of brakes, especially in heavy vehicular transports, could facilitate Cd insertions into road dust by frequent braking[68,69]. The Cd has a cluster at close distance with Ni and Cu but comparatively high accumulation and enrichment in road dust of RR and MuR, which can be associated with the frequent use of brakes of vehicles, particularly during traffic jams at peak hours. The concentrations of As, Cu, Hg, Mo, and Pb for the present study can coincide with previously reported PTMs from road dust of a mega city in China[70]. The Pb pollution can be attributed to the density of vehicular traffic on the roads and residual remains of historical usage of Pb containing fuels and lubricants. Though, historically efforts have been made to limit the Pb containing oils, paints, and lubricants for vehicular transportation but the density of traffic, especially on MuR and CR with elevated pollution levels of Pb[71,72]. With moderate pollution predominantly at MuR, WR, and JR, Pb strongly correlated with Zn and Co, pointing to vehicular emissions as a key source. High traffic from light vehicles in institutional areas contributes to brake wear, tire dust, and exhaust emissions, intensifying the accumulation of these metals in road dust. The high levels of enrichment and accumulation of Hg and Mo in the road dust suggest the association of its sources with roadside activities, especially in road dust of MaR, JR, and CR, which are considered institutional and commercial hubs, with dense traffic and numerous parking lots, including older, poorly maintained vehicles, contributing to the wear and tear of automotive components. This wear releases Hg and Mo from electrical systems and components, especially in the numerous vehicular maintenance workshops nearby. The use of Hg for various purposes in automobiles has become increasingly popular in recent years. For instance, every electrical connection in modern vehicles is activated by silver-coated connections. The Power windows, power seat adjustments, a power trunk, and important safety features are all operated by the silver membrane switches. The effective characteristics, such as low corrosive attributes, long-term usability, and high and safe conductivity of electric current, may be the cause of mercury enrichment and accumulation into road dust[73]. Based on the field characteristics, vehicular maintenance workshops are almost situated in every third corner of the local city areas, which subsequently contributing Hg pollution into the environment[74,75]. Besides, the presence of molybdenum in road dust may also be attributed to various aspects of the manufacturing processes of automobiles. For instance, molybdenum is subsequently used in steel for the fabricating of engine blocks, exhaust systems, crankshafts, piston rings, steering parts, and axel shafts. Molybdenum helps steel become stronger, harder, weldable, tougher, and more corrosion resistant. Furthermore, the low quality of usage and durability of vehicles with high chances of improper vehicular maintenance, sanitary and hygienic management make their accumulation more facilitated into the environment[27].

Health risk posed by PTMs
The findings of the present study concluded that potential health risks to children from the PTMs exposure in road dust of Lahore was noteworthy, especially Hg through ingestion. The HQing of Hg were most in MaR, which were 7.5 times higher for children than adults, showing mammoth concerns, whereas other selected PTMs showed no potential health risks. As and Cr were also indicating threatening levels of HQing, though less than the safe limit. The bioavailability of PTMs especially As and Cr mainly dependent on the oxidative state, for instance, Cr(III) is less toxic than Cr(IV). The toxicity of Cr(III) can metabolize lipid proteins and sugars in humans, whilst toxicity of Cr(IV) can cause cell mutation and prove to be cancerous to living organisms[76]. Also, the concentrations and variation of PTMs pollution levels in road dust are largely contingent upon the sampling time and/or seasons, which likely to influence the bioavailability and potential health risks on the population[77]. Earlier studies suggested the cruciality of PTMs exposure and noncarcinogenic risks in children through ingestion pathways because children are highly prone to ingest dust due to their higher hand to mouth activities; additionally, children have higher respiratory rate unknowingly in the dusty environment and dose to body weight ration with respect to identical dose exposure[27,70,78]. Therefore, limited exposure of children to the dusty environment is strongly recommended to protect children from adverse health impacts of PTMs. Additionally, frequent washing and cleaning of the roads would be helpful to dust remain deposited and to avoid the intact with humans.

Conclusion
Present study provides comprehensive insights into the current distribution, pollution status, and source identification of PTMs along with ecological and health risk assessment posed by PTMs in dust collected from eight prominent roads in Lahore, Pakistan. Results from pollution indices and health risk assessments were remarkably consistent across all selected roads. Concentrations of Cd, Hg, and Mo exceeded international guidelines several times over. The EF and I_{geo} calculations revealed moderate pollution of As, Cr, Cu, and Zn; slight pollution of Pb and Sb; moderate to high pollution of Cd; and extreme pollution of Hg and Mo. These findings were echoed in the MIPI and MIRI calculations, where Cd, Hg, and Mo posed the highest ecological risks. Statistical analyses highlighted various groupings among PTMs, suggesting industrial, vehicular, and non-exhaustive vehicular sources of their pollution in urban road dust. Factors such as the intensive use of old model vehicles, high vehicular density, overcrowded roads during peak hours, and numerous vehicular maintenance workshops significantly contribute to PTMs presence in Lahore's road dust. Additionally, the high usage of Hg and Mo in manufacturing automobile spare parts, coupled with poor quality and inadequate maintenance, exacerbates pollution levels. Notably, the presence of Hg in road dust poses potential health risks in children compared to other PTMs, emphasizing the need for public awareness and policy interventions to reduce and control PTM pollution. Efforts by policymakers to enact regulations aimed at mitigating PTM pollution, particularly in cities of high socio-economic importance like Lahore, are imperative.

Data availability
The supporting data has been provided as supplementary material.

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Author contributions

Dr. Abdul Rehman and Dr. Shan Zhong, both considered first authors, participated in the conception, design, methodology, data visualization, data interpretation, and writing the original draft. Dr. Xiaojun Zheng and Professor Daolin Du supervised the preparation of this article and finalized the final version of the article along with funding and resource acquisition. Ms. Samra Ijaz, Mr. Muhammad Irtaza Sajjad Haider, and Dr. Mudassar Hussain participated in experimentation, formal analysis, data interpretation, visualization, and revising it critically for important intellectual content.

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Declarations

Competing interests

The authors declare no competing interests.

Additional information

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