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Size-Segregated Elemental Profile and Associated Health Risk Assessment of Road Dust along Major Traffic Corridors in Kolkata Mega City

Deepanjan Majumdar ^{1,*}, Bratisha Biswas ², Dipanjali Majumdar ¹ and Rupam Ray ²

¹ Kolkata Zonal Centre, CSIR-National Environmental Engineering Research Institute, i-8, Sector C, EKDP, EM Bypass, Kolkata 700107, India; ds_majumdar@neeri.res.in

² Asutosh College, University of Calcutta, 92 Shyamaprasad Mukherjee Road, Kolkata 700026, India; bratishabiswas@gmail.com (B.B.); rupamray14nov@gmail.com (R.R.)

* Correspondence: d_majumdar@neeri.res.in; Tel.: +91-33-24415999; Fax: +91-33-24417608

Abstract: Particle size distribution (PSD) of road dust has significant repercussions on atmospheric pollution by road dust resuspension. The PSD of road dust at a few major commercial, traffic, and residential sites in Kolkata mega city was analyzed in the size range of <28–2000 μm . Predominance of the coarse size range (212–600 μm followed by 106–212 μm) was observed. In size-segregated road dust, Fe (4.02–31.2 g kg^{-1}) dominated other elements and was followed by Mg (2.13–10.9 g kg^{-1}), Mn (79.2–601 mg kg^{-1}), Li (395.8–506.8 mg kg^{-1}), and others. Fine particles (<28 μm) had higher elemental concentrations than coarser ones. Cd and Li showed the highest degree of enrichment compared to the Earth's crust, but only Cd posed significant ecological risk due to its high ecological toxicity. Individual elements did not pose significant non-cancer health risks, except for Li in children. However, the cumulative non-cancer risk from all toxic elements for children was almost four times higher than the acceptable level. Lifetime exposure to carcinogenic elements at the current level may pose 5 to 6 times higher cancer risk in the adult population than the acceptable risk of one in a million.

Keywords: air pollution; dry sieving; dust resuspension; human health risk; exposure; optical analysis; street dust



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1. Introduction

Road dust is a complex mixture of particles of both natural and anthropogenic origins. The former is derived primarily from soil, plant, and animal kingdoms (e.g., mold spores, animal dander, pollen, pollen fragments) and atmospheric deposition. The latter comes from construction and demolition materials (asphalt, concrete, paint), road wear and tear, automobiles (tire and brake wear and tear, body rust, tailpipe exhaust, etc.), and industrial inputs [1–4].

Road dust shares a dynamic relationship with the ground-level atmosphere and gets resuspended in the air via the sweeping action of wind and vehicle movements on roads [5,6]. In a resuspension chamber study, Martuzevicius et al. [7] found that $\text{PM}_{2.5}$, PM_{10} , and PM_{total} increased proportionally with increased airspeed from 8 m s^{-1} to 15 m s^{-1} . Dust particles with an aerodynamic diameter of about $<1 \mu\text{m}$ to about $100 \mu\text{m}$ may become airborne, depending on their origin, physical characteristics, and ambient conditions [8]. The Urban Air Quality Management Strategy in Asia (URBAIR), based on the estimates of $\text{PM}_{10}/\text{TSP}$ ratios for different sources, found that 20% of atmospheric PM_{10} came from road dust resuspension [9]. In Barcelona (Spain), Amato et al. [10] estimated that road dust accounted for 17% in PM_{10} , 8% of $\text{PM}_{2.5}$, and 2% of PM_{1} , implying that resuspension was responsible for 37%, 15%, and 3% of total traffic emissions, respectively, in PM_{10} , $\text{PM}_{2.5}$, and PM_{1} . It was estimated that annual total suspended particulates (TSP)

emissions accounted for about 240,000 t y⁻¹ in 2000 in urban and suburban Beijing. Traffic resuspended dust contributed more than 30% of atmospheric TSP [11]. In Kanpur (India), road dust emission was a significant source of air pollution, amounting to 25–50% of tailpipe emissions [12].

Particle size distribution (PSD) of road dust is of extreme importance as it governs mobility of particles in the association of attached pollutants and, together, govern air pollution potential of an area by resuspension [13,14]. In a study in Haidian District, Beijing, China, the median diameter of road dust (d_{50}) ranged from 100 to 200 μm [1]. Road dust from selected land-use types were predominantly fine particles (<250 μm , 60–75%), while grain size of 62–106 μm was the most abundant amongst eight size fractions, contributing $27 \pm 5\%$ and $38 \pm 21\%$ from college & residential and main traffic roads, respectively. Zafra et al. [15] reported that 82% of road dust vacuumed directly from the road surface was <1000 μm and 6.5% was <63 μm in Torrelavega (Northern Spain), while dust collected after sweeping was much finer, in which about 98% was of <1000 μm and 27% was under 63 μm . Vaze and Chiew [16] reported 10% and 15% contribution of vacuumed and swept road dust particles, respectively, in <100 μm road dust particles. Smaller particles with lower density, greater surface area per unit volume [17], and potential association with higher amounts of organics and pollutants are of particular concern [18,19]. A significant positive association between PM_{2.5} in road dust and hospital admissions due to cardiovascular and respiratory complications have been reported [20,21]. Therefore, road dust has immense importance from environmental and health perspectives. Populations with specific occupations such as drivers, roadside hawkers, shop owners, workers in roadside offices, and traffic police personnel are particularly vulnerable due to their occupational exposure towards fine road dust and road dust-borne contaminants.

Kolkata is a megacity and one of the largest in the world in terms of population density of 24,252 persons per sq. km, underlining the importance of likely health risk from road dust resuspension [22]. In a recent study, it was reported that the only significant risk combination (hazard index) in Kolkata was lead (Pb) exposure to children via road dust and ingestion was the dominant risk pathway [23]. ADB [24] had reported that about 15% of the average contribution (ranging from 5% to 28% over various seasons) of road dust atmospheric PM_{2.5} and about 60% of respirable particulate matter were contributed by road dust in Kolkata during 2003. A recent emission inventory study for Kolkata indicates that vehicle-induced road dust resuspension contributed about 15 kt y⁻¹ of PM₁₀ in 2015, i.e., 25% of the total estimated PM₁₀ emission. Due to increasing vehicular movements, road dust is projected to contribute 41 kt yr⁻¹, i.e., about 48% of the total PM₁₀ in 2030 [25]. Proper control of road dust may reduce the PM₁₀ and PM_{2.5} load of the air of Kolkata city by 7.5% and 2.3%, respectively [26].

Investigating the PSD of road dust vis a vis potential elemental signature at busy city locations is crucial to assess spatiotemporal variation in road dust and potential health risks. The PSD of road dust in Kolkata city has not been studied in detail yet. This study was designed to examine PSD of road dust at some significant commercial and densely populated areas in Kolkata, along with elemental loading in size-segregated road dust, keeping in view the potential effects of dust exposure on human health.

2. Materials and Methods

2.1. Background of the Study Area and Road Dust Sampling

The present population of the Kolkata Municipality area is more than 4.5 million, making it the third-largest city after Delhi and Mumbai in South Asia [22]. Due to issues like agglomeration, congested narrow roads, and construction and repairing activities, the city experiences substantial air pollution by particulates and is one of the non-attainment cities in India in terms of air pollution [27]. High emissions from the firing of smoking fuels in commercial eateries, use of adulterated fuels in two- and three-wheelers, lack of maintenance of vehicles, heavy traffic, congestion at traffic intersections, road encroachment

by pavement dwellers and street hawkers, etc., lead to substantial air pollution in the city [28].

Locations selected for road dust collection were mainly busy traffic roads, feeder roads, and service roads made up of asphalt coats. Most of the chosen sites were lined by shops and other commercial establishments, while a few were near bus stands, petrol pumps, and other government offices (Table 1). The sites were classified into traffic, commercial, residential, traffic + commercial, and traffic + residential areas based on primary activities or major inhabitation types observed around the sites. The sampling sites are depicted in a map vis-a-vis the city roads and urban agglomerations (Figure 1). Road dust was collected from about 3–4 points over an approximately 8 m² area on asphalt city roads and turned into one composite sample per site. Road dust was collected using the ‘Brush and Pan’ method, which is the most common method of road dust collection reported in about 88% of reviewed studies worldwide that collected bulk road dust samples ($n = 177$) [29]. Road dust was collected by thorough brushing of deposited dust, in a way so as not to cause abrasion on the road surface, by an inert nylon brush, and then stored in sample containers after collection in a stainless steel pan. Thorough brushing was done on the sampling spots to ensure maximum recovery of all deposited particles, including fines. Gravels, leaves, visible fibers, broken twigs, if any, large construction materials, and other large particles (>3 mm) were discarded during collection. A total of 11 site-specific samples of road dust were collected.

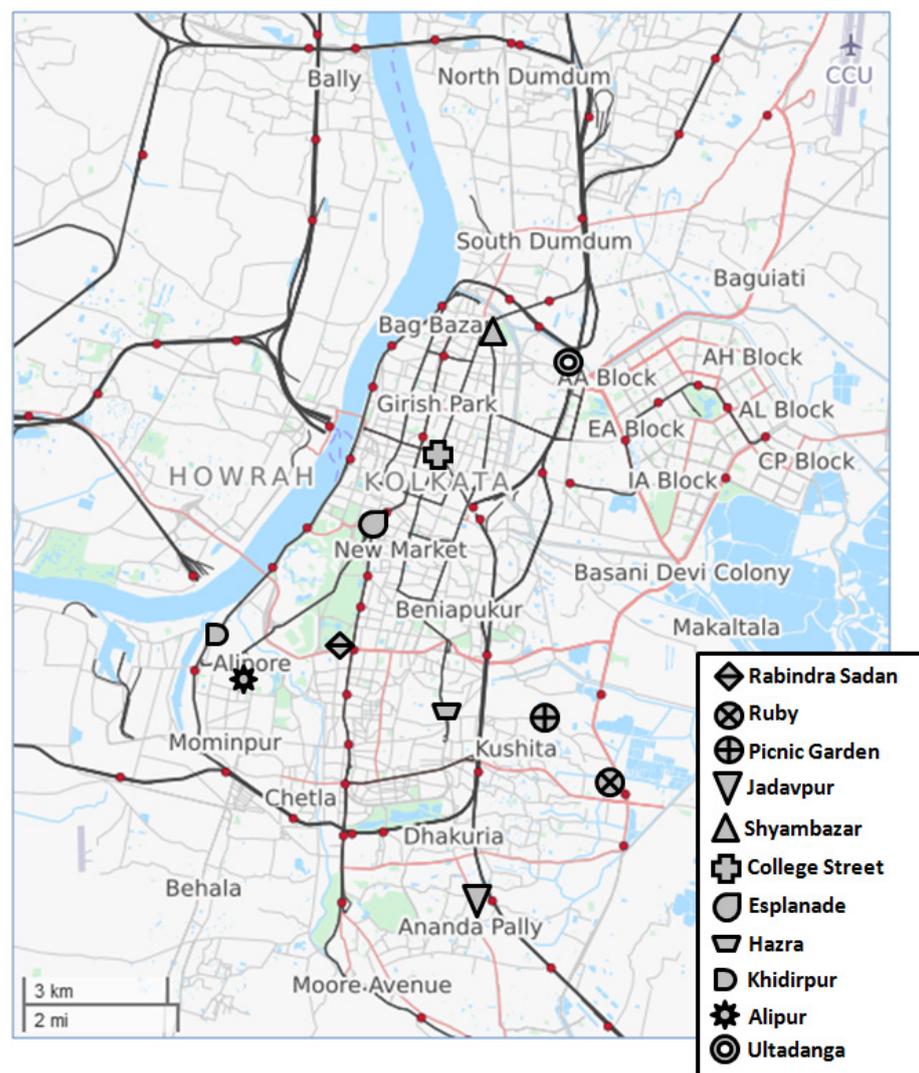


Figure 1. Road dust collection sites in Kolkata City (major road transport corridors are marked as dotted black bands).

Table 1. Summary of road dust collection sites.

Site Name	Site Coordinates	Details
Commercial Area		
Alipore	88.3363° N 22.5268° E	Semi-congested area; Asphalt Road; Court and Urban Local Body office nearby, a bus stop and a petrol pump are nearby
Residential Area		
Jadavpur	88.3707° N 22.4940° E	Asphalt Road; Low vehicular load; Railway station is within 500 m; A flyover is just adjacent; Local market is nearby
Picnic Garden	88.3884° N 22.5266° E	Semi-congested; Dotted by residences and small roads; Low vehicular road; Busy traffic square is nearby
Traffic Area		
College Street	88.36408° N 22.5774° E	Congested Area; Asphalt Road; High vehicular traffic; Presidency College and College Square are nearby
Ruby Square	88.4029° N 22.5135° E	Congested area; Asphalt Road; Heavy construction activity; Very high vehicular traffic; Gateway Hotel, a petrol pump are nearby; Small roadside food stalls that use biomass cookstoves
Ultadanga	88.3402° N 22.5927° E	Asphalt Road; Very high vehicular traffic; Circular rail station is nearby
Traffic + Commercial Area		
Rabindra Sadan	88.3451° N 22.5433° E	Wide Asphalt Road; Heavy vehicular traffic; Cinema Hall and Childrens' Museum are nearby
Hazra	88.34706° N 22.52372° E	Congested Area; Asphalt Road; Heavy vehicular traffic; Cancer Hospital, College and a big Park are nearby
Esplanade	88.3504° N 22.5647° E	Wide asphalt road; Heavy vehicular traffic; Mosque, a metro station, Income Tax Office, etc. are nearby; Large stores are also there in the vicinity
Shyambazar	88.3731° N 22.6006° E	Congested area; Asphalt Road; Very high vehicular traffic; Metro station is nearby; High commercial activity and surrounded by food stalls
Khidirpur	88.3268° N 22.5404° E	Congested Area; Asphalt Road; Traffic load is high; Commercial area, a bridge and a large market, etc. are nearby

2.2. Particle Size Distribution Analysis

Particle size distribution (PSD) analysis of road dust samples was undertaken using two different methods. A microcontroller-based electromagnetic sieve shaker (EMS-8, Electrolab, Electrolab India Pvt. Ltd., Navi Mumbai, India), having tri-dimensional movement integrating a vertical movement with a rotation, was used to segregate and determine PSD of road dust samples in 5 different cut-off sizes viz. > 2000 µm, 1000–2000 µm, 600–1000 µm, 212–600 µm, 106–212 µm, 63,106 µm, 45–63 µm, 28–45 µm, and ≤28 µm. Recovery of road dust samples from the series of sieves ranged from 98.1–98.8%. Each size portion of dust samples was weighed and stored in a refrigerator. Percentage distribution of particulates in each size range was calculated and recorded.

Segregated road dust samples in <106 µm size range was subjected to finer particle size distribution analysis in an Optical Particle Size Analyser (make: Sympatec, Clausthal-Zellerfeld, Germany) fitted with a HELOS (Helium-Neon-Laser for Optical Spectrometry) sensor in dry sample feed mode. Within each HELOS, the primary physical diffraction set-up is realized by deploying a parallel laser beam that yielded an optimum optical alignment to analyze extended spatial arrangements of particles. The analyzer was operated with a pressure of 3 bar, a vacuum of 10 mbar, a feed rate of 50%, a temperature control of 10–60 ± 0.1 °C in a measurement duration of 5 s.

2.3. Analysis of Elements

Analysis of elements in road dust was restricted to <106 µm particles due to higher health risks associated with fine particles amenable to resuspension in air [30–33]. Sieved and oven-dried samples (105 °C for 2 h before analysis; about 0.1 g) were digested with 10 mL concentrated HNO₃ in a microwave digester (Start MOD, Make M/s Milestone s.r.l., Sorisole, Italy). After digestion, samples were cooled and diluted to 50 mL by ultrapure water (MiliQ, Make M/s Millipore) and filtered through a 0.2 µm PTFE filter (M/s Millipore). Altogether, fourteen elements (Cd, Cr, Co, Pb, Mn, Ni, Sr, Zn, Fe, Mg, Li, Ti, Cu, Ba) were determined in road dust using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES, make: Teledyne Leeman Labs). A multi-point external calibration curve was prepared to estimate elements in extracted samples from a multi-element standard (M/s Accustandard) through serial dilutions. Calibration curves were prepared in two concentration ranges, the lower in µg L⁻¹ range and higher one in mg L⁻¹ range. Extracted samples were first analyzed against µg L⁻¹ range and elements with concentrations beyond the µg L⁻¹ range were again analyzed against the mg L⁻¹ range calibration curve. No contamination was detected in reagent blanks.

2.4. Assessment of Elemental Pollution by Enrichment Factor and Other Indices

The enrichment factor (EF) approach was utilized to assess the degree of elemental pollution [34]. EF is defined mathematically as:

$$EF = \frac{\left(\frac{X_n}{R}\right)_{\text{dust}}}{\left(\frac{X_n}{R}\right)_{\text{background}}}$$

where, $(X_n/R)_{\text{dust}}$ and $(X_n/R)_{\text{background}}$ are the concentration ratios of element n and the normalizer R in the road dust and background material, respectively. Titanium (Ti) was used as the normalizer and average elemental concentrations in earth crust were used as background [35]. Ti was used as the reference element for geochemical normalization as: (1) Ti is naturally associated with earth crust, is stable, non-reactive, and inert standard element with respect to the physico-chemical parameters of the environment [36]; (2) Ti geochemistry is similar to many other trace elements; and (3) its crustal concentration is fairly stable and is negligibly added by anthropogenic activities which is an important criterion [37]. Degree of element pollution was classified into five categories [38]: (1) EF < 2 (depletion to minimal); (2) EF = 2–5 (moderate); (3) EF = 5–20 (significant); (4) EF = 20–40 (very high); and (5) EF > 40 (extremely high).

The Contamination Factor (C_f^i) provides a single pollution index of a given element and is quantified as the ratio of an element to the background concentration of the corresponding element [39]. The C_f^i is the ratio obtained by dividing the concentration of each element in road dust by the background values in the earth crust [35].

$$C_f^i = \frac{C_{\text{element}}}{C_{\text{background}}}$$

C_{element} and $C_{\text{background}}$ are the measured concentrations of the element 'i' and the background concentrations, respectively. In this study, the average crustal concentration for elements [35] was used as a reference concentration. The degree of contamination (C_{deg}) is the sum of the contamination factors of measured elements that indicates the overall and extent of contamination of the study sites. It is estimated as follows [40].

$$C_{\text{deg}} = \sum_{i=1}^n C_f^i$$

The C_{deg} of the contamination may be classified based the scale ranging from <8 to >32 with classes such as <8 , $8-16$, $16-32$, and >32 indicates low degree, moderate, considerate, and a very high degree of contamination, respectively [39,41].

The potential ecological risk index (RI), proposed by Hakanson [42], was calculated to assess the likely ecological risk levels of selected elements Hg, As, Cu, Cr, Zn, and Pb in four different fractions (<28 , $>28-45$, $>45-63$, and $>63-106 \mu\text{m}$) in road dust. RI was calculated using the following relationships [42,43].

$$RI = \sum_{i=1}^n E_i$$

where,

$$E_i = T_i f_i$$

$$f_i = C_i / B_i$$

where E_i is the potential ecological risk factor of element i , T_i is the element toxic factor, i.e., Cu = 5, Pb = 5, Cr = 2, Ni = 5, Cd = 30, and Zn = 1 [42,43], f_i is the element pollution factor of element i , which equals to the amount of element i in the sample (C_i) divided by its reference value (B_i) in earth crust. Levels of potential ecological risk assessment are <150 , $150 \leq RI < 300$, $300 \leq RI < 600$, and $RI > 600$ indicating low, moderate, severe, and serious ecological pollution level, respectively [42,44,45].

2.5. Human Health Risk Assessment

The population in an urban area is exposed to road dust daily. Exposure to the elements in the road dust can be potentially toxic and may pose considerable non-cancer and cancer health risks to the population of Kolkata. Pathways of human exposure to road dust can be ingestion (I_g), inhalation (I_h), or dermatological contact (D_m). The exposure (E) from chronic daily intake of potentially toxic elements (PTEs) through the above pathways has been calculated as per the following equations.

$$E_{ig} = \frac{C_i \times 10^6 \times R_{I_g} \times F_{exp} \times ED}{(BW_{avg} \times T_{avg})} \quad (1)$$

$$E_{ih} = \frac{C_i \times R_{I_h} \times F_{exp} \times ED}{(EF_p \times BW_{avg} \times T_{avg})} \quad (2)$$

$$E_{Dm} = \frac{C_i \times 10^6 \times SAF \times AF_{Dm} \times A_{skin} \times F_{exp} \times ED}{(BW_{avg} \times T_{avg})} \quad (3)$$

Cancer and Non-Cancer Risk Assessment

Integrated lifetime cancer risk or CR is estimated from Equation (4):

$$CR_i = E_{i(Ig)} \times SF_i \quad (4)$$

The non-cancer health hazard from exposure to PTEs has been estimated as hazard quotient, HQ as per Equation (5):

$$HQ = E_{i(Ih/Ig/Dm)} / RfD_i \quad (5)$$

RfD_i is a chronic exposure reference dose for PTE 'i', below which undesirable health complications are not expected.

The cumulative non-cancer health hazard from cumulative exposure to PTE 'i' is expressed as hazard index, HI, as per Equation (6):

$$HI = \sum_i HQ \quad (6)$$

The definition of various parameters and assumed values are presented in Table 2. [46–53].

Table 2. Definition of parameters and values for health risk assessment.

Parameter	Unit	Abbreviat-Ion	Assumptions for Health Risk Assessment
Ingestion Exposure	-	E_{Ig}	-
Inhalation Exposure	-	E_{Ih}	-
Dermatological Exposure	-	E_{Dm}	-
Observed concentration of element 'I' in road dust	-	C_i	-
Ingestion Rate	$mg\ day^{-1}$	R_{Ig}	Adult: 100; Children: 200
Inhalation rate	$m^3\ day^{-1}$	R_{Ih}	20
Frequency of exposure	$Days\ year^{-1}$	F_{exp}	365
Exposure duration	Years	ED	Adult: 24; Children: 6
Average body weight	Kg	BW_{avg}	Adult: 60 kg; Children: 18 kg
Averaging time	Days	T_{avg}	$(ED \times F_{exp})$
Particle Emission Factor	$m^3\ kg^{-1}$	EF_P	1.36×10^9
skin adherence factor	$mg\ cm^{-2}$	SAF	Adult: 0.07; Children: 0.2
dermal absorption factor	-	AF_{Dm}	0.001
Area of skin	cm^{-2}	A_{skin}	Adult: 5700; Children: 2800
Carcinogenic Slope Factor of element 'I'	$(mg\ kg^{-1}\ day^{-1})^{-1}$	SF_i	*
reference dose for chronic exposure of element 'i'	$(mg\ kg^{-1}\ day^{-1})$	RfD_i	*

Average values adopted from published literature [48–57]. * Oral SF and RfD values of different exposure pathways for individual toxic elements are given in Section 3.4).

The chronic exposure reference dose and risk values for PTEs are adapted from Risk Assessment Information System of USEPA and other published literature [53,54]. PTEs with established RfDs were only selected for calculating the health risk index.

2.6. Statistical Analyses

To categorize the sites in clusters based on similarity in particle size distribution (<28–2000 μm and <106 μm size ranges analyzed by dry sieving and optical analysis, respectively). Cluster analysis was undertaken on particle size distribution data by Ward Method in Statistica Software (Dell Software, Round Rock, TX, USA, Version 13). Further, the average size distribution for all sites in <28–2000 μm and <106 size ranges of road dust particles were made to undergo distribution fitting separately via CDF (Cumulative Distribution Function) plot method by Statistica Software (Dell Software, Version 13). CDF plots display theoretical CDF of fitted distributions and empirical CDF based on sample data to determine data fitness to distributions. In the generated plots, N designates the number of non-missing observations.

3. Results and Discussion

3.1. Physical Attributes of Road Dust

PSD analysis of road dust by dry sieving technique revealed a predominance of 106–600 μm particles in all the samples in which the 212–600 μm size group had the highest share, followed by 106–212 and 63–106 μm size groups (Supplementary Figure S1). In Jadavpur, about 57.5% of road dust belonged to 212–600 μm diameter, followed by Esplanade (52.4%), and then Alipore, College Street Rabindra Sadan (48.0–49.6%). But at Khidirpur, the share of 212–600 μm particles was lower than the 106–212 and 63–106 μm size ranges, only 20%, the lowest amongst all. At Khidirpur, 106–212 μm particles had the highest share (about 23%), which was very closely followed by 212–600 and 63–106 μm ranges (19% and 20.5%, respectively). According to USDA (United States Department of Agriculture) classification, particles having a diameter in the range of 250–2000 μm (0.25–2.0 mm) are mainly medium and coarse sand particles, while particles from 50–250 μm (0.05 to 0.25 mm) range are various fine sand categories [55]. Particles of 1–2 mm diameter, categorized as very coarse sand, had a maximum share of about 6.8% at

Picnic Garden followed by Ultadanga, while the percentage of gravel (>2 mm) was highest at Picnic Garden (4%) but generally low in all others (0.05–2.4%). The average share of finer particles in the ranges of 45–63 μm , 28–45 μm , and <28 μm was highest at Khidirpur (8%), followed by Picnic Garden (5%), while the average share at all other sites was <5%. The above results implied the dominant presence of particles that matched size ranges designated to various types of sand.

More refined PSD analysis of segregated road dust (<106 μm) by optical particle size analysis went down to the estimation of the <4.5 μm size range, showcasing particles of concern from an air pollution perspective. The criteria air pollutant, PM_{10} (particles with <10 μm aerodynamic diameter), matched most closely to <11 μm range (PM_{11}) measured by the optical analyzer. This size group was most dominant at Picnic Garden (~23.4% of the 0–106 μm particle range), followed by College Street (18% of the 0–106 μm particle range), Khidirpur (~16% of the 0–106 μm particle range), Hazra and Esplanade (both ~14.3% of the 0–106 μm particle range) and others (Supplementary Table S1). About 50% of inhalable, thoracic, and respirable groups of dust designated for work environment corresponds to 100, 10, and 4 μm particles, respectively [56]. In the reported size groups in this study, the nearest corresponding size groups to the above groups were PM_{106} , PM_{11} , and $\text{PM}_{4.5}$. The actual share of the other criteria particulate air pollutants, i.e., $\text{PM}_{2.5}$ (particles with <2.5 μm aerodynamic diameter), could not be revealed by the optical PSD analysis. The nearest particle size group assessed was <4.5 μm . The $\text{PM}_{4.5}$ had the highest share at Picnic Garden (10.42%), followed by College Street, Khidirpur, Hazra (8.73%, 6.9%, and 6.2%, respectively) while the other sites had similar shares (4.5–5.7%). The non-cumulative particle size distribution resembles a normal distribution with negative skewness in <4.5–106 μm size range (Figure 2). Zafra et al. (2011) [15] reported that road dust and sediments collected from drains, bicycle lanes exhibited positively skewed log-normal distribution. Similar particle size distribution was also reported in road and gutter surface dust [57,58]. The optical size analysis 10, 16, 50, 84, 90, and 99 percentile contributions are given in Supplementary Figure S2. Picnic Garden had higher percentiles of smaller particles in every size range, indicating the finer nature of this road dust. Other critical physical characteristics like Volumetric Mean Diameter (VMD), Sauter Mean Diameter (SMD), surface area to volume ratio (S_v), and specific surface area (S_m) are also reported (Supplementary Table S2). Road dust at Picnic Garden had the highest S_v and S_m (0.51 and 1877.3) and correspondingly lowest VMD and SMD (32.58 and 11.79) that confirmed the finer nature of road dust at this site. The S_v , S_m , VMD, and SMD ranges were 0.28–0.44, 1038.2–1617, 35.46–48.3, and 13.69–21.32, respectively, at other sites.

The average particle size distribution was tested for distribution fitting by cumulative distribution function (CDF) plots in <28 μm –2 mm and <106 μm size ranges. It was found that the data did not fit very well to a normal distribution (Supplementary Figure S3a,b). Cluster analysis was performed amongst sites, separately for <28–2000 μm and <106 μm size ranges and site-wise cluster trees were developed to categorize sites with similar PSDs. The height of vertical lines in the branching dendrogram signifies the degree of difference between branches; the longer the line, more significant is the difference. In <28 μm –2000 μm , the branching dendrogram represented similarity amongst the site-groups of Khidirpur and Picnic Garden, Shyambazar and Hazra; Rabindra Sadan and Ruby Square; and Esplanade and Alipore. Khidirpur and Picnic Garden group was much different from other groups. Sites of Ultadanga, College Street, and Jadavpur each were outstanding (Figure 3a). In <106 μm size, Rabindra Sadan and Ruby Square; Shyambazar and Jadavpur; Hazra and Esplanade; Picnic Garden and College Street could be categorized as similar sites. The Picnic Garden and College Street group differed from the other three groups (Figure 3b).

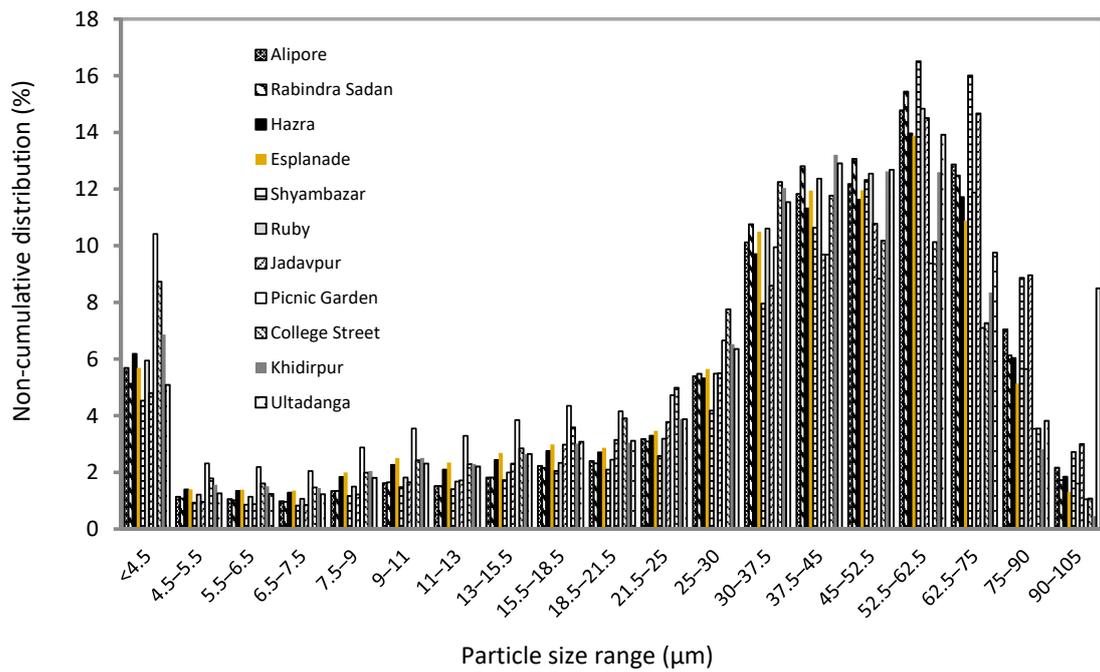


Figure 2. Non-cumulative distribution of road-dust (<106 μm size range) collected from different sites in Kolkata mega city.

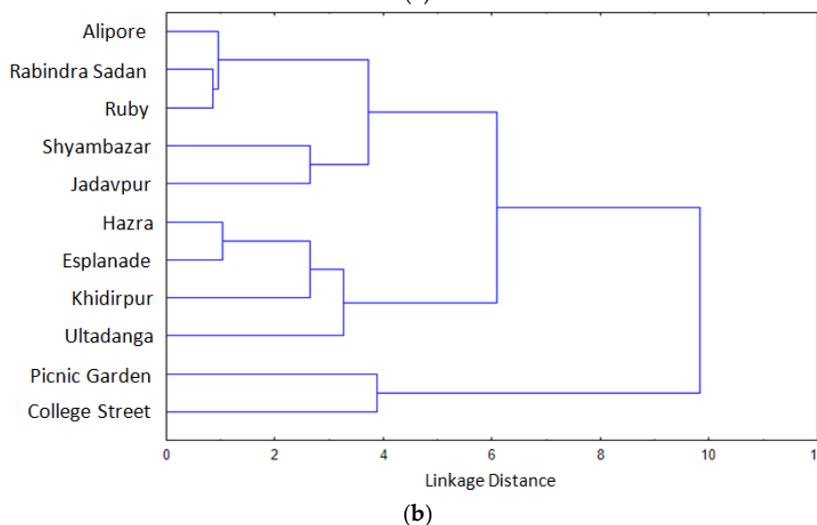
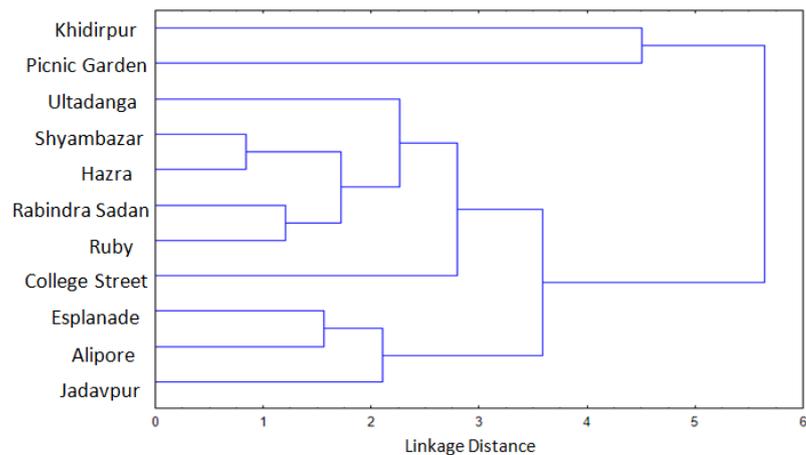


Figure 3. Tree clustering diagram depicting clustering of sites as per similarity in PSD of (a) road dust (<28–2000 μm size range) and (b) road dust (<106 μm size range).

3.2. Elemental Concentration in Road Dust

In size segregated road dust, Fe ($4.02\text{--}31.2\text{ g kg}^{-1}$) dominated other elements, including alkaline earth elements like Mg ($2.13\text{--}10.9\text{ g kg}^{-1}$), Mn ($79.2\text{--}601\text{ mg kg}^{-1}$), Li ($395.8\text{--}506.8\text{ mg kg}^{-1}$), followed by other detected elements above LOQ. The substantial presence of Li in Kolkata road dust may have to do with the presence of the sea at the Bay of Bengal within about 120 km from the city, implying the possibility of historical deposition of sea salt with Li on city roads, considering seawater is rich in Li [59]. Amongst the criteria elements (As, Ni, and Pb) earmarked in National Ambient Air Quality Standard (NAAQS) in India [60], Arsenic (As) was not detected in any sample. However, Ni and Pb were detected and ranged from $0.97\text{--}4.97$ and $14.16\text{--}67.11\text{ mg kg}^{-1}$, respectively, in various size groups (Supplementary Table S3). Amongst known toxic elements, Cr ($8.1\text{--}143.9\text{ mg kg}^{-1}$) recorded the maximum concentration, followed by Sr ($9.2\text{--}66.3\text{ mg kg}^{-1}$), Cd ($0.83\text{--}5.1\text{ mg kg}^{-1}$), Pb ($0.97\text{--}6.15\text{ mg kg}^{-1}$), and Co ($2.44\text{--}11.0\text{ mg kg}^{-1}$). Fe and Mg had a major share in total elemental load in all size groups ranging from $62.5\%\text{--}64.9\%$ and $27.6\%\text{--}29.3\%$, followed by Ti, Li, Mn, Ba, and so on (Figure 4). In an earlier study at Kolkata city [61], Cd, Cr, Cu, Ni, Pb, and Zn in road dust were $3.12, 54, 44, 42, 536,$ and 159 mg kg^{-1} , respectively, in road dust of the $<600\text{ }\mu\text{m}$ size range. In comparison, a recent study in the same city reported Fe, Cr, Mn, Co, Ni, Cu, Zn, Ba, Cd, and Pb concentrations in the ranges of $23.4\text{--}59.3\text{ g kg}^{-1}, 42\text{--}129, 503\text{--}1027, 8\text{--}18, 18\text{--}75, 28\text{--}279, 121\text{--}1258, 374\text{--}1643, 0.28\text{--}8.03,$ and $77\text{--}551\text{ mg kg}^{-1}$ in $<53\text{ }\mu\text{m}$ road dust. In road dust in Bengaluru city in India, the same elements were reported in similar ranges ($16.1\text{--}33.2\text{ g kg}^{-1}, 25\text{--}134, 258\text{--}621, 2\text{--}20, 9\text{--}192, 9\text{--}168, 43\text{--}617, 431\text{--}1921, 0.09\text{--}1.26,$ and $26\text{--}97\text{ mg kg}^{-1}$, respectively) [23]. In a study on the presence of elements in road dust in Delhi, India, Cd, Cr, Cu, Ni, Pb, and Zn in $<75\text{ }\mu\text{m}$ road dust were found to be $2.65, 148.8, 191.7, 36.4, 120.7,$ and 284.5 mg kg^{-1} [4]. The same study cited Indian soil background values as $0.9, 114, 56.5, 27.7, 13.1, 22.1$ for Cd, Cr, Cu, Ni, Pb, and Zn, respectively [62,63].

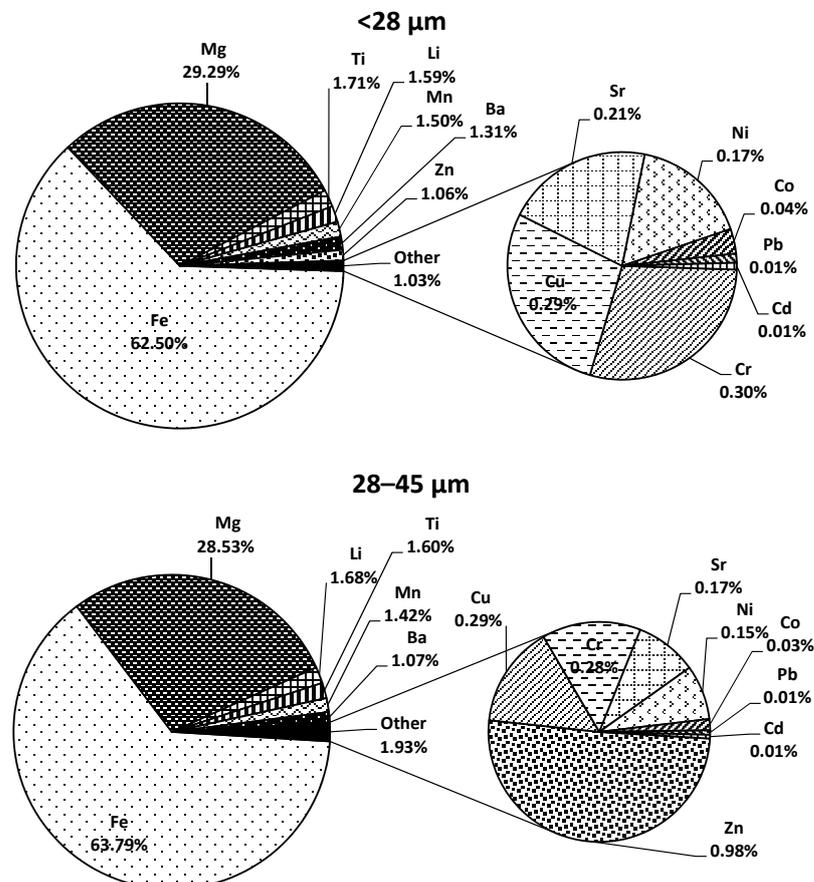


Figure 4. Cont.

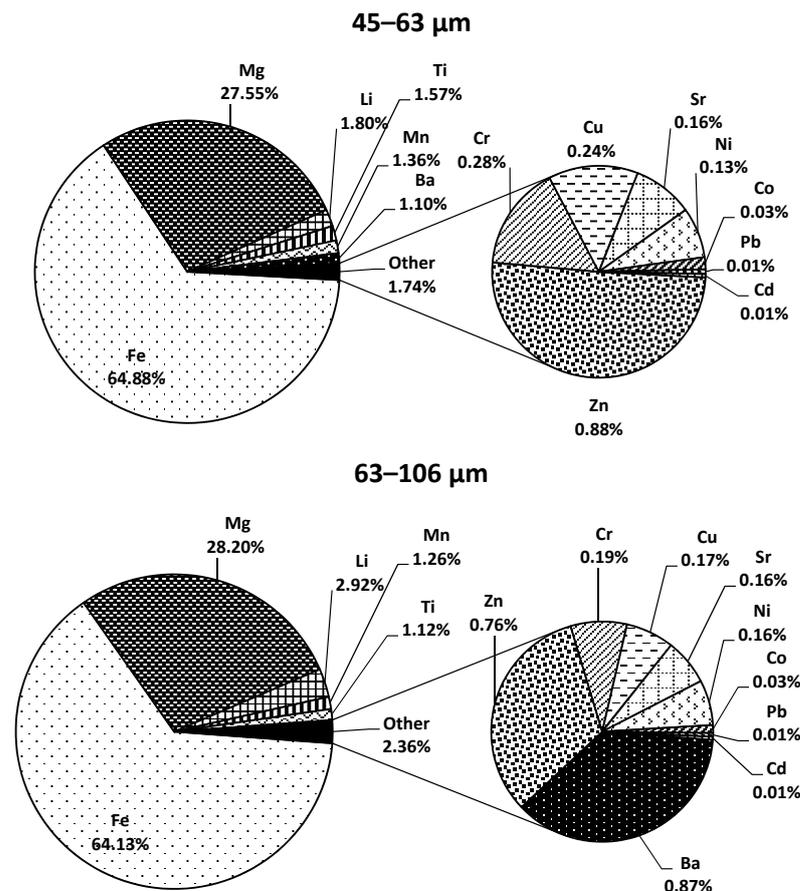


Figure 4. Percentage share of elements in road dust of various size groups.

With the exception of a few cases, <28 μm size group had a highest average concentration of the elements than larger size groups of >28–<45 μm, 45–<63 μm, and 63–<106 μm, while no wide variation in element concentration was apparent within each size group, as indicated by the coefficient of variation, CV (Table 3). Interestingly, the concentration of almost all elements except Li declined linearly, albeit to variable extents, with increasing particle size (Figure 5), indicating a higher association of elements in finer particles. The decreasing trends showed reasonably steep negative slopes with relatively high R² values (Supplementary Table S4). A substantial part of fine road dust is expected to get suspended in the air due to wind and vehicle movements [8]. Therefore, it is prudent to assess the potential ecological and health damages from likely exposure to elements attached to the finer dust.

Table 3. Average elemental concentration (mg kg⁻¹) in size-segregated road dust in Kolkata.

Parameter	Cd	Cr	Co	Pb	Mn	Ni	Sr	Zn	Fe	Mg	Li	Ti	Cu	Ba
<28 μm	2.68	71.27	9.11	3.53	402.19	38.85	66.32	297.16	14,736.23	8102.92	433.91	475.50	55.66	305.29
SD	0.47	17.49	0.72	0.63	66.02	7.89	4.45	45.02	3543.82	1304.46	20.32	76.35	44.34	78.11
CV (%)	17.62	24.54	7.85	17.74	16.42	20.30	6.71	15.15	24.05	16.10	4.68	16.06	79.65	25.59
>28–<45 μm	2.14	52.62	7.40	2.78	343.83	26.45	49.54	235.03	13,935.24	7380.53	452.82	420.64	38.63	225.50
SD	1.09	33.88	1.05	1.23	107.70	12.61	5.21	73.91	6170.33	1396.06	15.83	59.27	70.12	90.84
CV (%)	50.88	64.39	14.20	44.42	31.32	47.68	10.52	31.45	44.28	18.92	3.50	14.09	181.50	40.28
45–<63 μm	2.24	58.05	9.59	2.91	343.22	40.39	43.62	260.43	15,111.55	7572.23	468.99	375.60	63.00	341.24
SD	0.65	33.76	1.42	0.88	99.34	13.85	6.49	64.95	6404.68	1545.65	13.01	80.23	54.95	86.33
CV (%)	29.23	58.16	14.78	30.22	28.95	34.28	14.87	24.94	42.38	20.41	2.77	21.36	87.23	25.30
63–<106 μm	1.45	30.21	3.47	1.74	237.62	15.37	11.89	94.82	10,965.18	2749.98	502.46	188.52	11.08	66.87
SD	0.70	17.71	1.68	0.86	93.94	12.16	11.94	71.02	5192.17	2406.81	14.08	74.65	41.09	57.35
CV (%)	48.43	58.62	48.51	49.51	39.53	79.12	100.43	74.90	47.35	87.52	2.80	39.60	370.88	85.77

N.B. Average concentration in earth crust is adopted from Taylor, 1964.

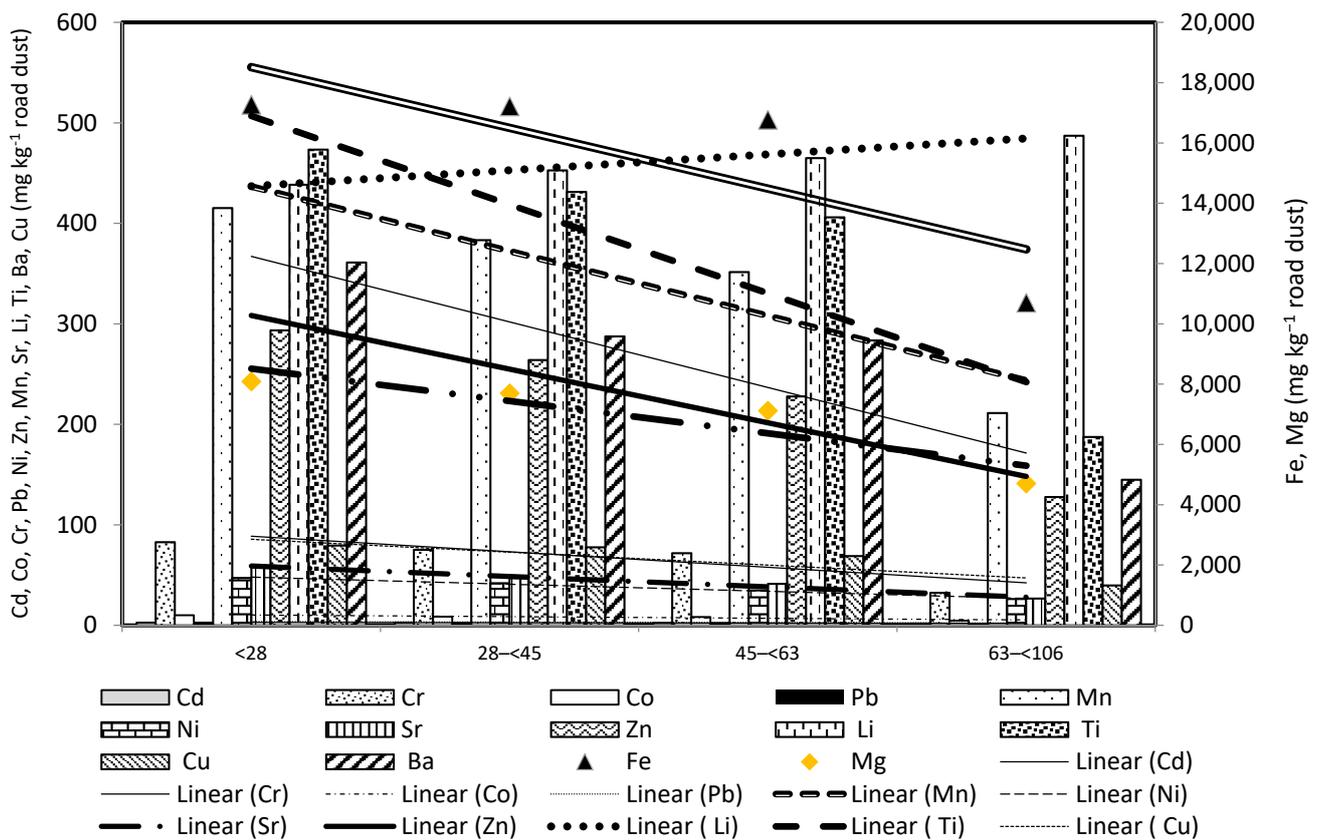


Figure 5. Decreasing trends in elemental concentration with increasing particle size.

3.3. Assessment of Elemental Pollution

The enrichment factors (EF) of elements in size segregated road dust revealed that Cd and Li had extremely high degree of enrichment. In contrast, the rest of the elements had significantly lower enrichment factors (Supplementary Table S5). Consequently, the degree of contamination in different size ranges was very high for Li (175 in <math><28\ \mu\text{m}</math> to 244 in $63-106\ \mu\text{m}$) followed by Cd (84 in $63-106\ \mu\text{m}$ to 109 in $28-43\ \mu\text{m}$). Amongst other elements, Zn and Cu showed a moderate degree of contamination (Table 4). The ecological risk was (Ei) estimated for six elements (Cu, Pb, Cr, Ni, Cd, and Zn). Cd posed the highest ecological risk values, ranging from 125 to 760 in different size ranges. The rest of the elements showed low ecological risk, ranging from 0.79 for Pb to 22.48 for Cu (Supplementary Table S6). It is evident from the potential ecological risk index (Ri) that only Cd posed a significant ecological risk, with Ri ranging from 2518 in $63-106\ \mu\text{m}$ to 3270 in $28-45\ \mu\text{m}$ size range (Table 5).

Table 4. Degree of contamination (C_{deg}) of metals in different size groups in road dust in Kolkata.

Metals	C_{deg}			
	<28	28–43	43–63	63–106
Cd	108.51	109.01	107.08	83.93
Cr	6.61	6.01	6.46	3.23
Co	3.21	2.76	2.99	1.82
Pb	2.37	2.19	2.30	1.73
Mn	3.50	3.23	3.33	2.22
Ni	5.03	4.46	4.17	3.48
Sr	1.26	1.00	0.99	0.71
Zn	33.54	<i>30.16</i>	<i>29.28</i>	<i>18.23</i>
Fe	2.45	2.45	2.68	1.90
Mg	2.78	2.64	2.75	2.02
Mn	2.30	2.24	2.26	1.31
Li	175.37	181.10	209.19	243.59
Ti	0.66	0.61	0.64	0.33
Cu	11.55	11.29	10.26	5.62
Ba	6.80	5.41	6.00	3.41

$C_{deg} < 8$ (low degree of contamination indicated by no highlight), 8–16 (moderate degree of contamination indicated by light grey highlight), 16–32 (*considerable* degree of contamination indicated by italics), and >32 (very high degree of contamination indicated by boldface).

Table 5. Potential Ecological Risk Index (Ri) of individual elements in various size groups in Kolkata.

Metal	Ecological Risk * in Particulate Size Range			
	<28	28–45	45–63	63–106
Cu	57.75	56.44	51.31	28.09
Pb	11.86	10.95	11.51	8.63
Cr	13.22	12.01	12.92	6.47
Ni	25.14	22.31	20.83	17.41
Cd	3255.15	3270.19	3212.35	2518.02
Zn	33.54	30.16	29.28	18.23

* Elements in a particular size range posing highly strong potential ecological risk are marked with boldface. Rest are of low potential risk.

3.4. Health Risk Assessment

Non-cancer health hazards from chronic element exposure through road dust to adults and children via ingestion, inhalation, and dermal contact pathways was assessed for twelve potentially toxic elements (PTEs, namely, Cd, Cr, Co, Pb, Mn, Ni, Sr, Zn, Fe, Li, Cu, and Ba) found in the road dust of Kolkata. These twelve elements have well-established non-cancer health impacts. Four elements (Cd, Cr, Pb, and Ni) have established carcinogenic health impacts and established slope factors for oral uptake. The cancer risk from long-term exposure to adults has been assessed for these elements for ingestion exposure only. This risk assessment was carried out assuming that the soil model applies to road dust as well. Complete assimilation of taken-up elements into the bloodstream was also assumed.

Co is the most toxic one among the studied elements, followed by Cd, Li, Cr, and Pb respectively, indicated by very low RfD values for ingestion exposure. Other elements are comparatively less toxic. Cd has the highest carcinogenic potential exhibited by the highest slope factor, followed by Ni, Cr, and Pb, respectively. The health risk assessment results are presented in Table 6. Non-cancer health risks are indicated by estimated Hazard Quotients (HQs) for different exposure pathways, and carcinogenic risk values are indicated as estimated Cancer Risk (CR) values. It was observed in the HQs that for all elements that the most significant exposure pathway was ingestion. Non-cancer health hazards for all elements were high for the ingestion pathway, followed by dermal contact and the lowest via inhalation pathway in adults and children alike. Similar estimates have been reported in other risk assessment studies from road dust exposure [50,53,64]. They reported HQ

from the ingestion pathway to be 10 and 100 times more than from dermal contact or the inhalation pathway respectively. The size range of settled road dust is generally $>10\ \mu\text{m}$. Here, elements in road dust of $<28\text{--}106\ \mu\text{m}$ size range were studied. Particulates of $>10\ \mu\text{m}$ diameter are filtered through our nasal follicles while breathing and cannot enter human lungs or subsequently in the bloodstream. The non-cancer risk of road dust is negligible for the inhalation pathways for both adults and children. Children have a higher exposure to road dust than adults, as they spend more time outdoors. Children also have a higher hand-to-mouth interaction, especially during playtime, leading to even higher exposure than adults. Moreover, children with lower body weight experience a considerably higher weight normalized exposure. Consequently, children face a higher risk of non-cancer health impact from exposure to PTEs in road dust. The dermal contact pathway of exposure to Fe for children was observed to be considerable. In this study, HQs corresponding to individual elements have not exceeded unity, indicating that they do not pose significant non-cancer health risks for adults or children, except for ingestion exposure to Li in the case of children. The same from exposure to Li can still be considered 'of concern' for adults, while exposure to Fe, Cr, and Co can be regarded as 'of concern' for children.

The cumulative HI from all PTEs was estimated to be less than unity (0.58). Therefore, exposure from target PTEs in road dust may not pose a significant non-cancer health risk for the city population. However, the same for children was assessed as 3.8, almost four times the desired HI of unity. Thus, it can be concluded that children are more vulnerable to the PTEs in road dust and have a significant probability of suffering from non-cancer health complications. Here, it was assumed that the HIs for different PTEs are additive, and the synergistic or antagonistic effect of cumulative PTE exposure, if any, has not been considered.

Among the four carcinogenic elements, Cd, Ni, and Cr posed a significant cancer risk in more than acceptable limits, i.e., one in a million for the exposed city population. This assessment indicates that an inhabitant of Kolkata city has a 5 to 6 times higher risk of developing cancer upon lifelong exposure to the city road dust. In Kolkata, 1.4 million people, i.e., 31% of the total population, resides in the city slums, most of which are settled along the roadside walkway or footpath [65]. Their daily chores, including cooking, sleeping, leisure, etc., occur just adjacent to the city roads. Therefore, they may have a much-elevated exposure to the road dust and PTEs compared to the rest of the city population living in better housing. In addition, children residing in slum settlements are also highly vulnerable to non-cancer health effects, primarily via the ingestion pathway. Therefore, children would have a higher chance of developing cancer upon lifelong exposure to road dust if they continue to live in roadside slums.

Table 6. Cancer and non-cancer health risk indicators from exposure to PTEs present in road dust.

Parameter	Exposure Type	Cd	Cr	Co	Pb	Mn	Ni	Sr	Zn	Fe	Li	Cu	Ba
RfD (mg kg ⁻¹ day ⁻¹)	Ingestion	1.0 × 10 ⁻³	3.0 × 10 ⁻³	3.0 × 10 ⁻⁴	3.5 × 10 ⁻³	1.4 × 10 ⁻¹	2.0 × 10 ⁻²	6.0 × 10 ⁻¹	3.0 × 10 ⁻¹	7.0 × 10 ⁻¹	2.0 × 10 ⁻³	4.0 × 10 ⁻²	2.0 × 10 ⁻¹
	Inhalation	-	2.8 × 10 ⁻⁵	6.0 × 10 ⁻⁶	3.5 × 10 ⁻³	5.0 × 10 ⁻⁵	2.6 × 10 ⁻²	-	3.0 × 10 ⁻¹	7.0 × 10 ⁻²	-	4.2 × 10 ⁻²	-
(mg kg ⁻¹ day ⁻¹) ⁻¹	Dermal	-	7.0 × 10 ⁻⁵	-	5.2 × 10 ⁻⁴	1.8 × 10 ⁻³	5.4 × 10 ⁻³	-	6.0 × 10 ⁻²	2.2 × 10 ⁻³	-	1.2 × 10 ⁻²	-
	SF	1.5 × 10 ¹	4.2 × 10 ⁻¹	-	8.5 × 10 ⁻³	-	9.1 × 10 ⁻¹	-	-	-	-	-	-
HQ (Ingestion)	Adult	4.1 × 10 ⁻³	3.6 × 10 ⁻²	4.5 × 10 ⁻²	1.5 × 10 ⁻³	4.3 × 10 ⁻³	3.0 × 10 ⁻³	1.2 × 10 ⁻⁴	1.3 × 10 ⁻³	3.8 × 10 ⁻²	3.8 × 10 ⁻¹	2.8 × 10 ⁻³	2.3 × 10 ⁻³
	Children	2.7 × 10 ⁻²	2.4 × 10 ⁻¹	3.0 × 10 ⁻¹	1.0 × 10 ⁻²	2.8 × 10 ⁻²	2.0 × 10 ⁻²	8.2 × 10 ⁻⁴	8.9 × 10 ⁻³	2.6 × 10 ⁻¹	2.6	1.9 × 10 ⁻²	1.5 × 10 ⁻²
HQ (Inhalation)	Adult	-	5.7 × 10 ⁻⁴	3.3 × 10 ⁻⁴	2.3 × 10 ⁻⁷	1.8 × 10 ⁻³	3.4 × 10 ⁻⁷	-	2.0 × 10 ⁻⁷	5.6 × 10 ⁻⁵	-	4.0 × 10 ⁻⁷	-
	Children	-	1.9 × 10 ⁻³	1.1 × 10 ⁻³	7.5 × 10 ⁻⁷	5.9 × 10 ⁻³	1.1 × 10 ⁻⁶	-	6.5 × 10 ⁻⁷	1.9 × 10 ⁻⁴	-	1.3 × 10 ⁻⁶	-
HQ (Dermal)	Adult	-	6.2 × 10 ⁻³	-	4.1 × 10 ⁻⁵	1.3 × 10 ⁻³	4.5 × 10 ⁻⁵	-	2.7 × 10 ⁻⁵	4.9 × 10 ⁻²	-	3.8 × 10 ⁻⁵	-
	Children	-	2.9 × 10 ⁻²	-	1.9 × 10 ⁻⁴	6.2 × 10 ⁻³	2.1 × 10 ⁻⁴	-	1.2 × 10 ⁻⁴	2.3 × 10 ⁻¹	-	1.8 × 10 ⁻⁴	-
HI	Adult	4.1 × 10 ⁻³	4.3 × 10 ⁻²	4.5 × 10 ⁻²	1.6 × 10 ⁻³	7.3 × 10 ⁻³	3.1 × 10 ⁻³	1.2 × 10 ⁻⁴	1.4 × 10 ⁻³	8.7 × 10 ⁻²	3.8 × 10 ⁻¹	2.9 × 10 ⁻³	2.3 × 10 ⁻³
	Children	2.7 × 10 ⁻²	2.7 × 10 ⁻¹	3.0 × 10 ⁻¹	1.0 × 10 ⁻²	4.0 × 10 ⁻²	2.0 × 10 ⁻²	8.2 × 10 ⁻⁴	9.0 × 10 ⁻³	4.8 × 10 ⁻¹	2.6	1.9 × 10 ⁻²	1.5 × 10 ⁻²
CR (oral) (10 ⁻⁶ population)	Adult	6.1 × 10 ¹	4.6 × 10 ¹	-	4.6 × 10 ⁻²	-	5.5 × 10 ¹	-	-	-	-	-	-

RfD—Reference Dose for chronic exposure; SF—Oral Slope Factor; HQ—non-cancer hazard quotient; HI—non-cancer hazard index; CR—cancer risk. NB: boldface values indicate significant health risk; values in *italics* indicate health risk 'of concern'.

4. Conclusions

The nature and characteristics of road dust of Kolkata city have been studied for size distribution and elemental composition in various size ranges. Road dust collected from important traffic corridors of the city reveals its typical urban characteristics, despite site-to-site variation. The predominance of coarse particles (106–212 μm) and a comparatively lower share of fine particulates (<28 μm) in Kolkata road dust may be considered advantageous from an air pollution perspective, as coarser road dust would possibly have a lower residence in air. Road dust showed a strong presence of coarse dust with about 83–98% in the sand particle size range. The characteristic size distribution of road dust varied across various parts of the city as indicated by cluster analysis. Fe and Mg are the two primary elements dominating the metal composition of road dust with 92% contribution. Li followed by Cd in the different size ranges of road dust showed the highest enrichment compared to their abundance in earth crust. Zn showed high enrichment in fine (<28 μm) fraction, but Cd constitutes only about 0.01% of the metal composition of the road dust. However, owing to its low concentration in the earth's crust and high ecological risk, Cd in road dust of Kolkata is a concern.

Potentially harmful elemental content in the road dust of the city may pose considerable human health risk upon chronic exposure at the prevailing levels. Ingestion was estimated to be the most significant pathway for exposure for both adults and children. Exposure to an individual element does not indicate significant non-cancer health risk, except for exposure to Li for children. The cumulative non-cancer health risk is also not indicated to be significant for adults. However, the same is about four times higher than the acceptable level, indicating that children are at risk of non-cancer health impact from chronic exposure to toxic elements of the city road dust. The city inhabitants have a 5 to 6 times higher risk of developing cancer upon lifelong exposure due to the current level of three carcinogenic elements, namely, Cd, Ni, and Cr, in road dust. A sizable number of city populations residing in the roadside slums, including children, are even more vulnerable to the health impact of road dust exposure.

Regular road sweeping and cleaning remains an important option for local urban bodies in Kolkata to ensure city roads' cleanliness. Kolkata Municipal Corporation (KMC) has street sweeping as one of its primary activities [66], which needs to be fortified in terms of coverage and frequency, especially in areas with possibilities of substantial human exposure vis a vis population density. Deployment of vacuum-assisted road sweeping machines may be a pragmatic way forward for effective cleaning of city streets.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/atmos12121677/s1>, Figure S1: Particle size distribution of road-dust samples (<28–>2000 μm size range) collected from different sites in Kolkata, Figure S2: Percentile of particle size distribution of road-dust (<106 μm) collected from different sites in Kolkata, Figure S3: Plot of empirical cumulative distribution function (CDF) of average particle size distribution of (a) road dust (<28–2000 m) (b) road dust (<106 m size range) against a normal distribution plot, Table S1: Summary of non-cumulative particle size distribution of road dust (<106 μm particle size range), Table S2: Volumetric mean diameter (VMD), Sauter Mean Diameter (SMD), surface area to volume ratio (S_v) and specific surface (S_m) area of road dust (<106 μm road dust), Table S3: Metal concentration (mg kg^{-1}) in size-segregated road dust at different sites in Kolkata, Table S4: Slope of decline in metal concentration with increasing particle size in size-segregated road dust, Table S5: Enrichment factors of metals and degree of pollution in road dust with respect to size range, Table S6: Range of Ecological Risk Values (E_i) of individual metals in different size groups in road dust of various sites in Kolkata.

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Majumdar, D.M. (Dipanjali Majumdar); visualization, D.M. (Deepanjan Majumdar); supervision, D.M. (Deepanjan Majumdar); project administration, D.M. (Deepanjan Majumdar). All authors have read and agreed to the published version of the manuscript.

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