



Heavy metals in urban road dusts from Kolkata and Bengaluru, India: implications for human health

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Abstract Air pollution and dust pollution are major urban environmental issues, with road dust being a potential source and a pathway for human exposure. The developing megacities of India, where the population may spend a significant portion of their working lives close to the roadside, including consuming street food, have obvious source–pathway–receptor linkages. The aim of this study in Kolkata and Bengaluru, India, was to evaluate the risk to human health from inorganic components of road dust. Samples were collected and analysed from a cross section of urban environments for a wide range of anthropogenic and geogenic elements, some such as antimony showing an increase in response to vehicle activity. Calculated enrichment factors relative to crustal abundance demonstrated significant enrichment in common heavy metals and less commonly reported elements, e.g. molybdenum, antimony, that

may be used as contaminant markers. Factor analysis gave multielement signatures associated with geography, vehicle traffic and local industry. The bio-accessibility of road dusts in terms of ingestion was determined using the BARGE method with more than 50% of zinc and lead being available in some cases. A formal human health risk assessment using the US EPA framework showed that lead in Kolkata and chromium in Bengaluru were the elements of most concern amongst chromium, nickel, copper, zinc and lead. However, the only risk combination (hazard index) shown to be significant was lead exposure to children in Kolkata. Ingestion dominated the risk pathways, being significantly greater than dermal and inhalation routes.

Keywords Road dust · Risk assessment · Kolkata · Bengaluru · Bio-accessibility

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Introduction

Air pollution and dust pollution are major urban environmental issues in many nations; rapidly developing megacities are particularly prevalent in South-east Asia with the movement of large numbers of people (receptors) to these urban areas in search of employment. Part of the rapid growth inevitably includes planning for industrial, road network and environmental health falling behind development, for

older cities such as Kolkata, India, compared to newer cities such as Bangalore, India. The result being road dust a source, pathway and sink for contamination could have a detrimental influence on a population living and working in close proximity. Road dust as a potential health issue has been investigated across many developing megacities (MCs), and examples in decreasing world rank (Cox 2019) that the current study can be compared against include Tokyo MegaCity (MC)1 (Kumar et al. 2013); Delhi MC3 (Chutke et al. 1994; Banerjee 2003); Mumbai MC6 (Kumar et al. 2001); Guangzhou MC11 (Huang et al. 2012); Beijing MC12 (Men et al. 2018a, b); Dhaka City MC13 (Ahmed and Ishiga 2006); and Kolkata MC19 (Chutke et al. 1994; Chatterjee and Banerjee 1999; Karar and Gupta 2006; Gupta et al. 2007).

Hazard to humans from road dust can be broadly divided into three categories: (1) mineralogical, where physical effects on pulmonary tissues are of concern (Checkoway et al. 1997), (2) organic chemicals, for instance poly-aromatic hydrocarbons (PAHs) (Lorenzi et al. 2011), and (3) inorganic species with potentially toxic elements (PTE) including Cr, Cu, Ni, Pb and Zn (Liu et al. 2014). When considering these hazards, risks and implications for human health, the *source–pathway–receptor* paradigm is useful in outlining any conceptual model. All three elements are required for a risk to be evident; however, the presence of all three does not imply an unacceptable risk. Unacceptable risk is usually the result of modelling based on real data. The use of this paradigm is commonly recommended as part of environmental legislation and human health guidance (SNIFFER 2007).

The hazards to health from (3) inorganic species in road dusts are the focus of the current study, and the paradigm may be broken down as follows:

Sources

In the urban environment, sources are generically well known and are often summarised into the following categories: (1) motor vehicle traffic (exhaust, tyre and brake wear products (Mummullage et al. 2016)); (2) associated road infrastructure [tarmac (Mummullage et al. 2016), degraded metal road furniture (Thorpe and Harrison 2008; Keuken et al. 2013)]; (3) industrial processes such as metal production, working and plating (Chatterjee and Banerjee 1999), plastics

manufacture and working (Turner 2016); and (4) power generation, e.g. coal, oil and gas (Chen et al. 2015). The importance of these sources will change from city to city depending on economic focus and individual sources in their scope for contamination; this may also vary from city-wide injection of fine particles and gases from power plants to very local, medium-sized dusty industry such as ceramic production (Querol et al. 2010) or small-scale light industry such as metal working. An additional issue is the role of time; firstly, sources may change over time, not just in terms of intensity of environmental inputs but also in terms of the formulation of contamination. If road dust is a sink for a contaminant and this material does not degrade into a less toxic form, contaminant concentrations may increase over time, even for a constant input rate. An extreme example of the role of time in risk assessment is leaded petrol, which although used worldwide in large quantities between 1950s and 1990s, has now been discontinued due to health risks particularly for children, but the lead dispersed into the surface environment is still potentially available and may be re-circulated, i.e. remobilisation of deposited dust, during storms or by fast moving vehicles.

Pathways

Heavy metals associated with road dust can have implications for human health via two main exposure pathways: inhalation into the lungs and ingestion via hand and mouth. The dermal exposure pathway was not considered likely to be significant for heavy metals in this study for the general population as they would not on a day-to-day basis handle road dust and contact would be limited to a small amount of material settling on exposed skin. This would only be significant for workers in specific job-related scenarios such as those sweeping the roads and pavements or those in car repair shops. Worldwide, the inhalation pathways are classically more often considered the most important as exemplified by the large volume of studies on PM10 fractions (Kong et al. 2011); the lower volume of literature considering the ingestion pathway may lead to a down-playing of its significance in urban population risk assessment. For Kolkata in particular, the ingestion route may be very significant due to the large number of street food vendors operating in areas of high traffic density. The application of the EPA risk

assessment for the different pathways is an opportunity to test this hypothesis. Direct *in vivo* bio-availability testing as part of risk assessment is rarely practicable; therefore, bio-accessibility as a proxy has been validated (Denys et al. 2012) and is therefore used in this study.

Receptors

Human receptors in Kolkata in common with large cities in many developing countries may spend a significant portion of their day on the street working and, in some cases, sleeping out in the open (*as observed in this study during sampling*).

The overarching aim of the current study was a human health risk assessment of heavy metals in road dust from two contrasting Indian megacities, Kolkata (MC19) and Bengaluru (MC32): the first is a historically industrialised city, whilst the latter is a modern major technology centre. Road dusts were collected and analysed for both potentially toxic elements (PTE) and matrix elements with the objectives of: (1) identifying those with significant anthropogenic enrichment; (2) source assignment; (3) the ingestion bio-accessibility of heavy metals in road dusts; and (4) the generation of hazard indices using standard models after (Khairy et al. 2011; Liu et al. 2014) giving a quantitative human health risk. Particular perspectives for these results were sought by comparison with other megacities and the relative importance of the ingestion compared to inhalation and dermal absorption.

Methodology

Geographical, climatic and socio-economic context of study areas

The chosen study sites were within the urban centres and surrounding suburban areas of Kolkata (MC19) and Bangalore (MC32), India (Electronic Appendix EA: Figure 1).

Kolkata (formerly Calcutta) is the capital and administrative centre for the state of West Bengal. A population of 14.1 million was officially recorded for 2011 (Census of India 2011), this being 4.3% of India's urban population and 1.3% of the total population (PNUD 2009), but this was estimated to have increased to 15.2 million (Cox 2019).

Kolkata has a tropical climate with a mean temperature of 26.2 °C, whilst the maximum temperature is c.30 °C from April till August and this falls to 20 °C in December and January. The mean annual rainfall is 1735 mm, whilst maximum rainfall occurs between June and Sept and the minimum between December and January (anon. 2019a); as a result of the monsoon, the city and West Bengal have the highest rainfall on the eastern coast of India.

Kolkata was the capital of India until a century ago and suffered an industrial turn-down from the 1950s. Since 2011, a more business-orientated state government has pushed economic development (Chaudhuri 2012). Kolkata has one of the most affordable public transport systems in India (anon. 2018), but the present extension of the underground railway system consists of buses, taxis and three-wheelers (auto-rickshaws), and a majority of these are old, diesel fuelled and highly polluting. Until the end of the last decade, the three wheelers used to run on “kata-tel”—a mixture of kerosene and used diesel, which although banned, can still found being used outside the city (Das et al. 2016).

Bengaluru (also known as Bangalore) is the capital and administrative centre for the state of Karnataka. A population of 8.5 million was officially recorded for 2011 (Census of India 2011), this being 2.0% of India's urban population and 0.6% of the total (PNUD 2009), but was estimated to have now reached 11.3 million (Cox 2019). Bengaluru has a tropical climate with a mean temperature of 23.6 °C, whilst the maximum temperature is c.27 °C from April to May and this falls to 21 °C in December and January. The annual rainfall is 831 mm whilst maximum rainfall occurs between September and October, the minima is in January and February (anon. 2019b).

Bengaluru is viewed as the engine of Indian technology growth and was ranked 12 in a world list of technology-rich cities above Delhi, Mumbai and Kolkata (anon. 2015). According to a recent report by the Central Pollution Control Board (CPCB), India, Bengaluru stands top of the six major polluting cities in India due to the large number of diesel engines and increasing industrial activities at the cities edge (Moudgal 2018). Other studies indicate that the groundwater around the industrial area in Bengaluru is contaminated by a number of heavy metals including Cr (Shankar 2009).

Sample site selection

The selection of sampling sites was based on obtaining a cross section of urban exposure environments across a significant and representative portion of each city. Therefore, sites chosen included road junctions with high and very high road vehicle density, these typically representing outdoor work/commercial/shopping environments; smaller side streets with medium and low road traffic density, dominated by more domestic dwellings; and paths and access roads within parks and open spaces that could be considered urban background sites, i.e. < 2500 vehicles/day within 50 m (Putaud et al. 2004). These sites and their “Relative Traffic Index” (RTI) are listed in Electronic Appendix EA: Table 1. The RTI was based on observing the relative number of vehicles during sample collection and should be considered a qualitative ranking rather than a quantitative measure. The geographical distribution of sample sites is shown in Fig. 1a (Kolkata), b (Bengaluru). Sites were mapped using open source software and data: Quantum Geographical Information System (QGIS), version 3.2.1-Bonn; the Indian country outline was obtained from <http://www.naturalearthdata.com>, whilst for city base-maps data were downloaded from <http://www.openstreetmap.org> and then key features were drawn using QGIS to give diagrammatic maps.

Sample collection

Individual road dust samples were collected using a simple plastic dust pan and brush approach. Areas swept included road or pavement surfaces and gutters across where possible at least 10 m² to ensure representative sampling. Particular attention was paid to material lodged in dust traps such as pavements cracks, as flat surfaces were frequently denuded of material due to wind and therefore only contained material representative of a short timescale, whereas the traps probably represented more time-averaged material. Collected material was carefully transferred from the pan to clear plastic bags for transport and storage to minimise losses of finest material. The dust pan and brush method despite its simplistic nature continues to be a regularly used option in road dust sampling (e.g. Men et al. 2018a)

A majority of sampling in Bangalore was conducted by one of the authors (MC) but in Kolkata a

number of different samplers were used. For health and safety reasons, samplers went out in pairs, allowing one to be a spotter whilst the other a collector. This also gave the opportunity to collect duplicate samples at sites each using a different sampler. This allowed an estimate of sampling uncertainty including both collection area and collector.

Sample preparation

All samples were shipped to the British Geological Survey (BGS) in the UK for preparation and analysis. On arrival, samples were air-dried (< 40 °C) and carefully nylon sieved to < 200 microns. This is the laboratories’ default for material used when testing materials for ingestion exposure, as suggested in (USEPA 2007). It should be noted, however, that (Juhasz et al. 2011) suggest this may underestimate Pb concentrations as smaller particle sizes with elevated concentrations may be preferentially transferred to the mouth via the hands. The sieved fraction is further milled to < 53 microns in an agate ball mill for homogenisation purposes prior to “total” analysis.

Sample analysis

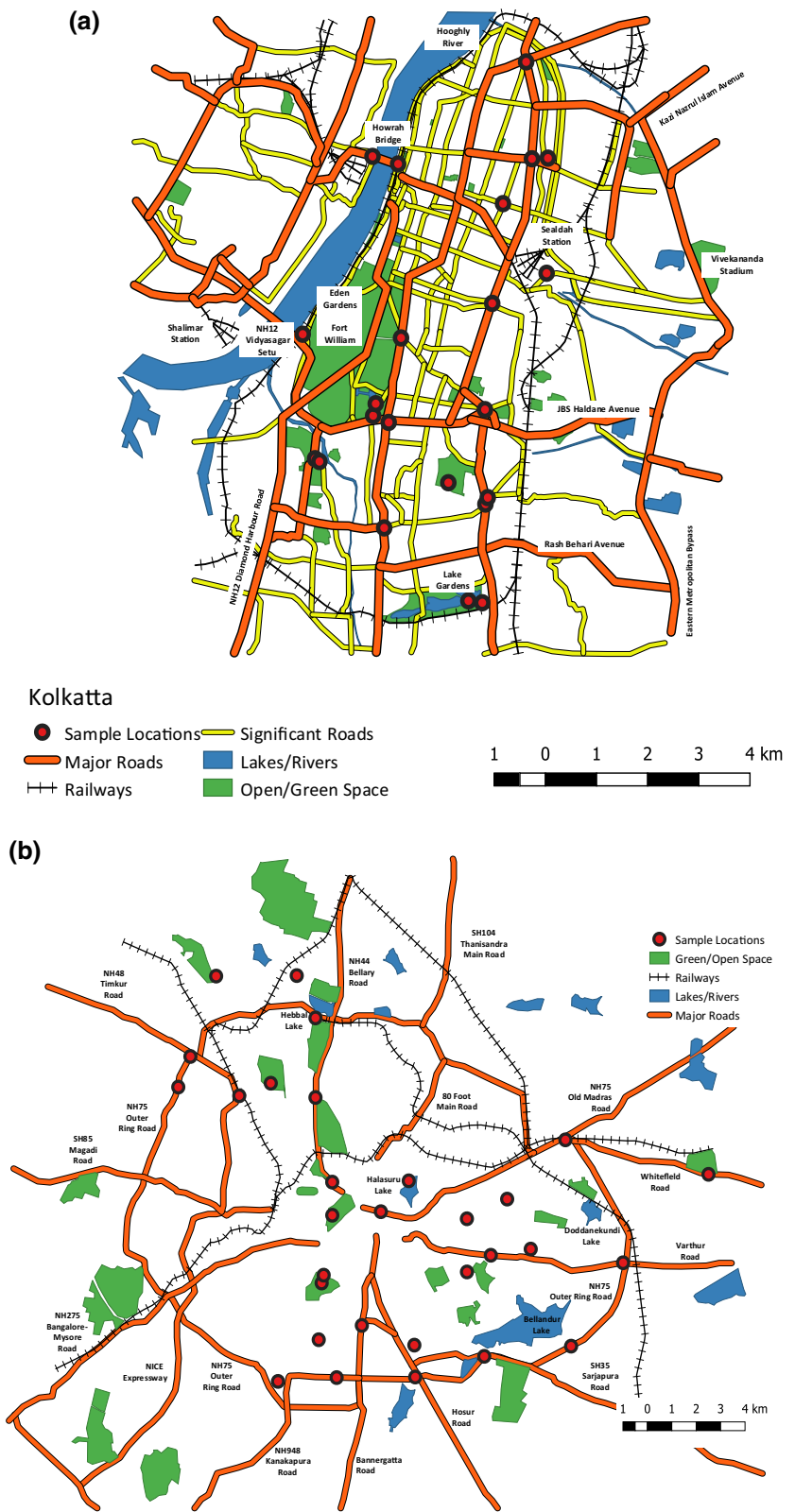
Total concentration determinations

The digestion procedure for total elemental determinations was based on an HF-based mixed acid attack (see Electronic Appendix: Method 1). In addition to samples, multiple reference materials, blanks and duplicate samples were prepared within the same batch.

All solutions were analysed by ICP-MS after appropriate dilution. Data were corrected for matrix suppression and blank contributions. Data quality were evaluated using chemical control solutions and the digested reference materials for accuracy and precision; duplicates for typical sample reproducibility and digestion blanks for total method detection limits. Away from the detection limit, typical accuracy and precision across the range of elements were better than 10% RSD.

Mercury analyses were performed separately using a direct Hg analyser (thermal desorption—atomic absorption spectrometer). Quality control was assured with solid certified reference materials and blanks.

Fig. 1 a Kolkata and **b** Bangalore. Distribution of sampling sites around cities. Base map from open source data on OpenStreetMap.com



Bio-accessibility test

The bio-accessibility test used in this study is referred to as the Unified BARGE Method (UBM), as described fully in (Denys et al. 2012; Hamilton et al. 2015). The UBM is an in vitro technique for simulating the human digestive process using a mixture of synthetic organic and inorganic fluids. The stomach (gastric)-only process uses a combination of saliva and gastric fluids, whereas the stomach intestinal process uses duodenal and bile simulants. Blanks, duplicates and reference materials (Hamilton et al. 2015) were analysed with each batch.

Data processing and statistical interpretation

All data processing was performed using Microsoft Office 16 ExcelTM, whilst statistical interpretation was performed using the Minitab Inc. MinitabTM (v17) statistical package.

Anthropogenic enrichment factors (EF)

Enrichment factors (EF) for each sample were calculated following the double normalisation methods (Atiemo et al. 2012; Chen et al. 2014), using Al as the normalising element and the average upper crustal concentrations (Rudnick and Gao 2003), with the degree of enrichment classification (Lu et al. 2010) where $2 < EF \leq 5$ moderate; $5 < EF \leq 20$ significant; $20 < EF \leq 40$ very high; and $EF > 40$ extreme.

Element: source assignment

The use of principal component analysis (PCA) and its derivative factor analysis (FA) to elucidate likely element–source associations is common, for example Gunawardana et al. (2012). Principal component analysis reduces the dimensionality whilst retaining the maximum information by using a linear combination of the dataset variables that are uncorrelated. Factor analysis also uses a linear combination of variables but with a user-defined number of latent factors, which may then be rotated such that simpler structure of loadings from the original variables is apparent, aiding interpretation. Prior to conducting this statistical analysis, data were conditioned in two stages. Firstly, to avoid outlier samples dominating, their concentrations were moderated. Values greater

than the robust mean +3 robust standard deviations were replaced with the robust mean +3 robust standard deviations. The second stage was to scale the data such that major element concentration and their associated variances did not dominate. There are two commonly used scaling techniques: (1) “z-scoring” where the mean is subtracted from each value before being divided by the standard deviation, e.g. Gunawardana et al. (2012), and (2) log transformation of the data. The second was chosen as much of the data were not normally distributed but tended to be a log-normal distribution. A number of elements were omitted from the FA, most due to high degrees of correlation, i.e. several of the rare-earth elements (REEs). The initial factor analysis demonstrated that the variance was dominated by the anthropogenic elements. Therefore, the elements in the factor analysis were reduced further to just those likely to be anthropogenically controlled, to focus on potentially different sources of contamination.

Results and discussion

Elemental concentrations in road dusts

The concentrations of 57 elements were determined in 34 road dusts from 23 sites in Kolkata and 30 road dusts from 30 sites in Bengaluru. The elements analysed consisted of both those with a geogenic source and those likely to be anthropogenically enhanced. Full elemental analyses are given in Electronic Appendix EA: Table 2.

To allow rapid evaluation of duplicate analyses across a large number of elements, a preferred difference between duplicates of < 10% was chosen, with a target difference of less than 20%. For the purposes of discussion below, this is limited to those anthropogenic elements of particular health concern (Cr, Ni, Cu, Zn, Pb and Hg) and example geogenic elements (Al, U) and Fe as a crossover geogenic/anthropogenic element. For the geogenic elements (Al and U) and the crossover element (Fe), either the preferred or target duplicate differences were achieved. The anthropogenic elements of health interest were more variable. Zinc and Pb, with single exceptions, reach the target difference. Copper and Hg regularly demonstrate differences between 20 and 100%; this may be partially explained for Hg by many

samples of very low concentrations (< 0.1 mg/kg). Nickel showed a range of differences between those observed for Zn–Pb and Cu–Hg. In attempting to understand this deviation from ideal, comparison of the difference observed between analytical duplicates and sampling duplicates suggests, as might be expected, the differences are typically slightly greater for sampling. However, the random nature of high differences in the analytical duplicates implies a “nugget” effect, despite milling and homogenisation. The obvious answer is to increase the milling time for future projects, but the efficacy of this potentially is limited by the tendency of metallic particles to smear rather than comminute. An alternative would be to increase the analytical sample size at an increased analytical cost. The final pragmatic alternative is to accommodate this potential variation by modelling using robust or nonparametric statistics and avoid over-interpreting relatively small absolute changes in concentration.

Histograms of the distributions of each elemental concentration are given in Electronic Appendix EA: Figure 2; a majority of elements show nearly normal or log-normal distributions with relatively few outliers. Normality and log normality were tested with the Shapiro–Wilk test and are given in Electronic Appendix EA: Table 3; only Ba, Ta and Bi failed both these tests for both cities, inspection of data suggests due to outliers. As a result, median rather than mean is the preferred method of comparison and data were all log-transformed before data investigation tools such as factor analysis. The range of data for Kolkata, Bengaluru and other comparable major cities is given in Table 1. The prevalence of each element defined as either geogenic or anthropogenic by city is summarised in Table 2; it is immediately clear from the major geogenic elements, e.g. Al, Ca, that there must be a fundamental difference in the base mineralogy between the cities and that Kolkata has typically higher concentrations of classic anthropogenic elements (Cu, Pb, Zn, etc.) but techno-anthropogenic elements such as Mo and W were similar between cities.

The control of traffic density on elemental concentrations was initially investigated by box and whisker plots of elemental concentration by City and RTI. A complete set of plots are given in Electronic Appendix EA: Figure 3. Elements that showed a marked concentration difference (smaller differences in

brackets) by RTI in Bengaluru included Na; (Mg); S; Ca; (Mn); (Fe); Cu; Sr; Mo; Sb; Ba; La; Ce; Pr; Nd; Sm; (Eu); (Gd); (Tb); (Dy), whilst in Kolkata elements showing a marked difference included (Ti); (V); Cr; Mn; Fe; Co; Zn; (Sb); and (Pb). These differences were judged by both an increase in interquartile range and median (Electronic Appendix EA: Figure 3). Noteworthy is the lack of a coherent set of elements, where concentration increased with RTI in both cities. The best example of a coherent element is Sb in Fig. 2.

The primary aim of the current study was the potential for an impact on human health from heavy metals in road dusts in two rapidly developing megacities in India. As the simplest measurable descriptor for this is the total elemental concentration, this is most easily put into context by comparison with other major developing cities. Table 1 directly compares where available, the range of values for a number of cities recorded in the literature and contains a single value, when only mean values were reported (Chatterjee and Banerjee 1999; Banerjee 2003; Ferreira-Baptista and De Miguel 2005; Ahmed and Ishiga 2006; Hu et al. 2011; Khairy et al. 2011; Singh 2011; Duong and Lee 2011; Huang et al. 2012; Atiemo et al. 2012; Kumar et al. 2013; Pathak et al. 2013; Du et al. 2013; Rajaram et al. 2014; Liu et al. 2015; Li et al. 2015; Herath et al. 2016; Padoan et al. 2017; Pan et al. 2017; Gope et al. 2017; Men et al. 2018b; Shi and Lu 2018; and Duan et al. 2018). The range was chosen for comparison as the most flexible metric, because in each study different types of sites were sampled, both directly impacted (by vehicles and industry) and urban background. Additionally, studies had varied methodologies including using various size-fraction cutoffs, digestion techniques and instrumentation for analytical measurements. Inspection of Table 1 shows that Kolkata and Bengaluru are typical cities in terms of elemental concentrations, e.g. Pb (mg kg^{-1}): Kolkata (77–551) and Bengaluru (26–97); minimum other cities 0, Colombo, Sri Lanka; maximum other cities 2452, Beijing, China.

Anthropogenic enrichment factors (EFs)

Summary statistics of EF by element and city are given for selected elements in Electronic Appendix EA: Table 4 and the complete analyte set in Electronic Appendix EA: Table 5. Figure 3 plots the median value for elements where one of the EF statistics

Table 1 A comparison of the range of elemental concentrations (mg kg^{-1}) in road dust from a selection of worldwide large and megacities as reported in the literature, with those reported in the current study for Kolkata and Bengaluru

City	References	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Mo	Cd	Sn	Ba	Pb	Hg
Kolkata	This study	46–104	42–129	503–1027	23,422–59,309	8–18	18–75	28–279	121–1258	2–16	0.4–17	0.28–8.03	5.0–17.0	374–1643	77–551	0.022–34
Bengaluru	This study	26–59	25–134	258–621	16,134–33,222	2–20	9–192	9–168	43–617	1–2	0.4–4	0.09–1.26	1.9–22.9	431–1921	26–97	0.004–0.098
Tokyo, Japan	Kumar et al. (2013)							442–1043	651–1043			0.51–0.91			44–220	
Shanghai, China	Shi and Lu (2018)	25–233					5–66	23–152	103–1025			0.19–3.66			14–192	
Beijing, China	Du et al. (2013)	40–115					14–45	16–623	79–533			0.2–2.0			29–2452	
	Men et al. (2018a)	52–145		396–860	21,221–55420		16–168	26–285	96–2197	2–15		0.18–1.80			22–261	0.04–0.78
Delhi, India	Banerjee, (2003)	0–9000					80–1000	0–1300	100–360			13–17			120–200	
	Pathak et al. (2013)	18–312	11–10,268	163–10,574	7119–349,670	2.5–524	18–443	15–2224	67–12884			0.3–11.7			22–190	
	Rajaram et al. (2014)	171	699	27,047		23.3	37	169	264					333	129	
Dhaka City, Bangladesh	Ahmed and Ishiga (2006)	53–95	63–172				19–49	7–215	50–240	4–9				192–2119	20–110	
Kolkata, India	Chatterjee and Banerjee (1999)	77	54	619	26,700	16	42	44	159	23	0.5	3.12	0.8	213	1030	1.95
Nanjing, China	Hu et al. (2011)	42–102	61–250	382–1294	17,900–66,000	7–21	25–268	29–272	140–798	10–19		0.44–2.2				0.05–0.34
	Liu et al. (2015)	67–391	67–391	527–1861	28,000–330,000		22–158	57–4237	120–720					383–2595	43–832	
Accra, Ghana	Atiemo et al. (2012)	171–4948	83–440	131–1270	1790–113,000			21–268	70–1840						25–815	
Asansol, India	Gope et al. (2017)							81–179	155–249			0.65–0.88			86–141	
Colombo, Sri Lanka	Herath et al. (2016)	71–489		452–1127	11,390–43,798			68–347	105–1657						0–371	
Alexandria, Egypt	Khairy et al. (2011)	39–102	59–149	398–568	21,531–38,540		25–59	56–242	236–8153			1.72–6.62	9.4–18.0	142–1818	174–710	

When a range was not reported, a single value is given; this is usually the arithmetic mean that is reported

Table 2 Prevalence of geogenic and anthropogenic elements by city as determined by histograms of elemental concentrations

	Bengaluru > Kolkata	Bengaluru ≡ Kolkata	Kolkata > Bengaluru
Geogenic Elements	Na, Al, K, Ga, Sr, Zr, Nb, La, Ce, Pr, Hf, Ta, Th, U	Li, Be, Rb, Ba, Nd, Sm, Gd, Tb, Er, Tm, Yb, Lu, Y	Mg, P, Ca, Ti, Se, Nb, Cs, Eu, Dy, Ho
Anthropogenic elements	Tl	Ni, Mo, Sb, W	S, V, Cr, Mn, Fe, Co, Cu, Zn, As, Ag, Cd, Sn, Pb, Bi, Hg

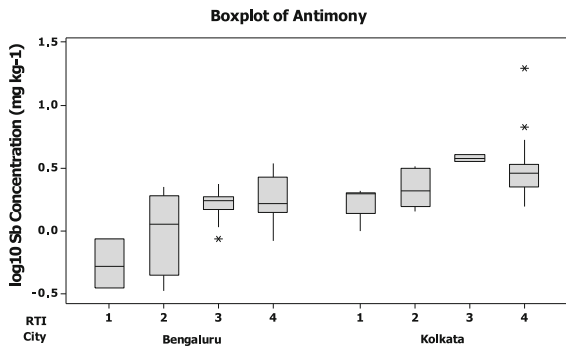


Fig. 2 Box and whisker plot of an example anthropogenic element, antimony (Sb) concentrations in road dusts changing by city and road traffic index (RTI). This is plotted as the log₁₀ of the concentration due to its log-normal distribution and large dynamic range

exceeded 2, i.e. moderate enrichment. Elements where none of the statistics reached this threshold included Li, Be, Na, Ti, V, Co, Ga, Rb, Sr, Y, Zr, Nb, Cs, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Hf, Tl. Also excluded from the table were Mg, K, Rb, Yb and Lu as they had only one or two samples which fell into the moderate category and are not normally considered harmful anthropogenic elements. A number of observations may be made from Fig. 3, EA: Table 4 and

EA: Table 5. Several elements not normally considered to be of anthropogenic origin are moderately enriched including Ca, Ta, Th and U, and this is likely associated with local geology, Kolkata being the city with higher values. Bengaluru is less enriched in elements traditionally associated with anthropogenic activities; only Ni, Cu, Zn, Sb, W reach significant or greater enrichment at their maximum values. In contrast, for Kolkata, the median values of Cu, Zn, Ag, Cd, Sn, Sb, Hg and Pb are all at significant enrichment or greater. However, Ag is the only element where the median enrichment value is very high or greater.

A comparison of previously reported enrichment factors for common heavy metals, from ten major cities, with those reported in the current study is made in Electronic Appendix EA: Table 6. These are normally reported relative to crustal abundance (Gope et al. 2017) or sometimes a local background uncontaminated soil or dust (Najmeddin et al. 2017). The normalising element varied with study, but the more common variation used was Al as in the current study. In a majority of cities (including Kolkata and Bengaluru) and for a majority of elements, the minimum enrichment factors were below the limit of EF > 5 for significant, high or extreme categories (Lu

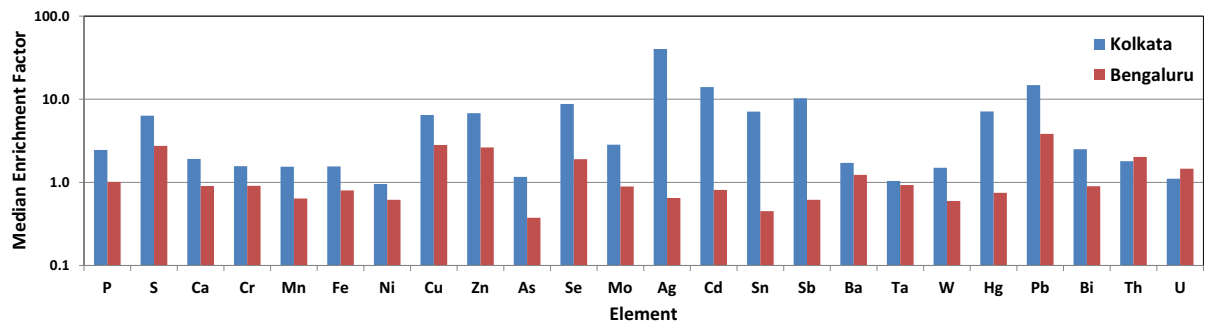


Fig. 3 Median enrichment factors (EF) for elements that show enrichment in one or more statistical measures, by city, relative to average continental crust for a selection of possible anthropogenic elements

et al. 2010). Perhaps of more interest is the maximum reported enrichment factor: *Cr*, only the Gold Coast, Australia, and Colombo, Sri Lanka, had significant or higher EF, with Kolkata and Bengaluru being similar to many others. *Manganese*, only London, UK, and Xi'an, China, have significant maximum EF, with Kolkata and Bengaluru being similar to many others. *Iron*, only six cities have reported data with London, UK, and Tehran, Iran, having significant EF, with Kolkata and Bengaluru being similar to others. *Nickel*, demonstrates for some metals the proportion of cities with significant maximum EF can be raised, i.e. 4 out of 7, with Bengaluru being one. *Copper*, *Zn* and *Pb* all commonly have maximum EF that is significant or higher: ten out of 12; nine out of ten and 11 out of 12, respectively, across most cities including Kolkata and Bengaluru. *Arsenic* was only reported in one other study (Xi'an) and Kolkata exceeds Xi'an but Bengaluru is lower in terms of maximum EF. *Cadmium* is unusual compared to the other elements as in Tehran, Changsha and Kolkata it reached an "extreme" level, whilst in Asansol, Gold Coast and Bengaluru, it is at most "moderate enrichment"; the extreme levels are presumably due to particularly dirty industries. Unfortunately, for several elements where in Kolkata (this study) we observed "very high" or "extreme" enrichments: Mo, Se, Ag, Sb, Hg and Bi, there are little or no comparable data available in the literature. It is worth noting that for Ahvaz, Iran (Najmeddin et al. 2017), an EF range for Sb was between 2 and 30 compared to Kolkata (4–74) and Bengaluru (0.2–6.5). *Antimony* is of particular interest due to its very low natural abundance and its use in brake linings and other fire retardants. It can thus be an excellent marker for vehicle activity (Fujiwara et al. 2011). Usefully, enrichment factors due to the double normalisation (both against average earth's crust and to a naturally occurring background element, i.e. Al) are inter-comparable between elements and therefore the extreme or very high enrichments of Cd, Mo, Se, Ag, Sb, Hg and Bi should make these good tracers of contaminant sources if sufficient data are available. Across all element enrichment factors and cities, it is clear that Bengaluru is one of those showing lower anthropogenic contamination, whilst Kolkata is extensively higher in anthropogenic input.

Element: source assignment

Factor analysis gives the element loadings associated with each factor and the proportion of the dataset variance associated with that factor (Electronic Appendix EA: Table 7). The closer the loading is to 1 or -1 , the greater the element's association with that factor, whereas a loading of 0 would imply no association with that factor. The advantage of factor analysis is an element with multiple sources will be split between factors associated with those sources and thus the relative importance of those factors (sources) may be evaluated.

Factor 1: dominated the analysis containing 58% of the total variance. All elements had very marked negative loadings except Tl, with the highest associations with Mn, Pb and Zn. *Factor 2*: 11% variance, was dominated by two contrasting groups of elements—those with a negative loading P, Ba, S, Hg, Bi, Tl, As and those with a positive loading Cr, Mo, Ni; the first group is chalcophilic and more volatile, and the second is associated with working metals or fossil fuel burning. *Factor 3*: 7% of variance; the positive loading is dominated by Cu and Sb, two elements associated with brake lining wear, and the negative loading by Tl, Co, Ni. *Factor 4*: 5% of variance; the elemental grouping did not show any obvious source significance. All other factors had a variance proportion of less than 5% and were not deemed of significant interest.

The individual sample factor scores were considered in terms of site and road traffic index to further elucidate possible sources, pathways or controls (data given in Electronic Appendix EA: Table 8). A simple bivariate plot of Factor 1 versus Factor 2 sample scores (Fig. 4) demonstrates that Factor 1 clearly separates the two cities, whilst for Factor 2 there is a much greater variability for Kolkata with respect to Bengaluru. Box plots of the Factor 1, 2 and 3 scores by city and RTI (Electronic Appendix EA: Figure 4) highlight the following. *Factor 1*: the negative factor scores associated with general high concentrations of anthropogenic elements are markedly lower in Kolkata and somewhat lower with increased RTI; lower values imply greater association. It should be noted that increasingly negative factor scores are not incompatible with increasing concentration of elements after the data processing. *Factor 2*: the negative loading ("steel"-type metals or fossil fuel burning) is more

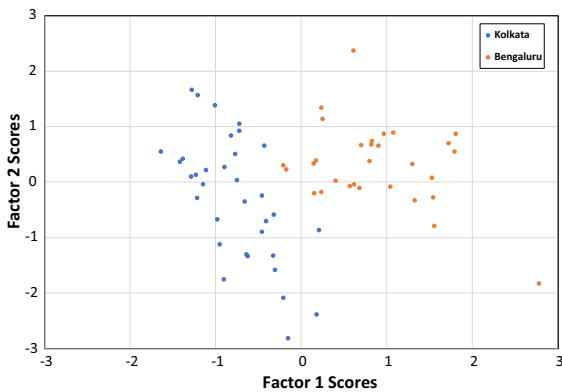


Fig. 4 Factor analysis of all Kolkata and Bengaluru samples. Bivariate plot of first two factor scores (58 and 11% of variance) for each sample showing a distinct separation by city. These factors are dominated by F1 (Mn, Pb, Zn negative loading and Tl positive loading) and F2 (P, Ba, S, Hg, Bi, Tl, As negative loading and Cr, Mo, Ni positive loading)

associated with Kolkata and markedly lower with at lower RTI. *Factor 3*: the sample scores are wider ranging for Bengaluru, and in both cities, the scores become more positive with higher RTI but to a greater extent in Bengaluru imply a greater influence of brake lining wear.

The use of PCA and its derivative factor analysis has been widespread in the literature as a means of identifying potential sources of contamination (Banerjee 2003; Atiemo et al. 2012; Chen et al. 2014) from multielement data sets. Often these have resulted in generic associations due to a lack of data on end-members. This was partially addressed by Mummullage et al. (2016) where road surface, tyre and brake lining material were also analysed. However, these data are inevitably limited by variations by manufacturers and country of production. The factor analysis elemental groupings and contextual information, i.e. RTI for the current study when compared to the literature, allow some degree of source apportionment. Factor 1 dominated by Mn, Pb, Zn but noteworthy for nearly all the contaminant elements in the current study is analogous to the general undifferentiated urban signal referred to by (Ferreira-Baptista and De Miguel 2005); whilst the use of Mn, an additive in unleaded petrol, has led to identifying a traffic factor in Santiago de Chile (Valdés et al. 2013), this is contradicted in this study by the lack of correlation between this factor and RTI and additionally Zn has been considered a nonspecific indicator for traffic-

related particulate matter due to its widespread use in metal industries (Keuken et al. 2013). Factor 1 highlights the markedly higher contaminant concentrations for most elements in Kolkata compared to Bengaluru and most probably reflects its much longer history of industrial processes, vehicular activity (particularly pre-removal of Pb from petrol) and ageing street furniture. Factor 2 is harder to equate with the literature from road dust studies; some of the two groups of elements (P, S, Hg, Bi, Tl, As; and Cr, Mo, Ni) may be associated with coal burning and oil/petrol burning, respectively (Klein and Russell 1973; Reddy et al. 2005; P. Chen et al. 2015). However, Mummullage et al. (2016) associate the second group of elements with tyre wear. This may account for the relationship in Kolkata between RTI and Factor 2 scores. Factor 3 has particular associations with Cu, Sb, Ba, all strongly associated with brake lining wear (Mummullage et al. 2016). In both cities, there is a good correlation between the factor scores and the RTI; this relationship is clearer in Bengaluru presumably as this has a lower background of other historical industrial sources. It is noteworthy that neither Factor 2 nor Factor 3 has any relationship with Zn, when many examples of its association with tyre wear (Atiemo et al. 2012; Mummullage et al. 2016) have been made; this necessarily introduces some caution into assigning factor 2 to vehicles. However, this may be explained if the general industrial signal in Factor 1 totally dominates the elemental variation across the two cities. Chatterjee and Banerjee (1999) associated Pb, As, Hg, Cd, Sn, Mn and Zn all with smelter activity in Kolkata.

Bio-accessibility of key health-related heavy metals

When considering the potential impact on health from the ingestion of dust, a knowledge of bio-availability/bio-accessibility is important as the use of total elemental concentrations may significantly overestimate the risk if these are low. In the current study, Cr, Ni, Cu, Zn, As and Pb were considered as key health-related heavy metals. In all cases except As, the stomach phase gave rise to higher bio-accessibility fractions (BAF) (Electronic Appendix EA: Table 9). When considering the median BAF, the order of accessibility was Zn > Pb > Cu > As > Ni > Cr. Perhaps of most concern is the relative availability

of Pb due to both its toxicity for children and the range of BAF (31–78%), this range is consistent with the full range (0.1–100%) observed in Denys et al. (2012) for exemplar contaminated soils. No elements showed a relationship between BAF and total metal concentration, suggesting that source material and the variable mineralogy of the dusts were a control. The exception was Zn for which there was an apparent nonlinear relationship (Electronic Appendix EA: Figure 5). This in part may reflect differences in the make-up of the dusts between Kolkata (BAF c. 55–85%) and Bengaluru (BAF c. 30–60%) and their small overlap in BAF. As many of the Kolkata samples show BAF approaching its maximum possible limit (100%), such a nonlinear trend is inevitable as a relatively accessible anthropogenic factor totally dominates a more refractory geogenic fraction with increasing concentration.

Human health risk assessment

The human health risk from road dust exposure was evaluated using Hazard Quotients (HQs) and Hazard Indices (HIs) derived using the US EPA framework (USEPA 2002), where $HQ = 1$ is the limit above which for a single variable and a single exposure route, a noncarcinogenic risk is likely to give adverse health effects (E. Liu et al. 2014). This framework considers the ingestion, inhalation and dermal exposure routes, modelling daily exposure doses for each route (CDI), where CDI accounts for the concentration in the road dust, the rate of ingestion or inhalation or skin absorption, exposure frequency and duration, body weight, average time of exposure and specifically for inhalation a particle emission factor, whilst for dermal exposure skin area, skin adherence and dermal absorption factor. This CDI is normalised to a reference dose (RfD) to give an exposure route hazard quotient (HQ), where the reference dose is the maximum value likely to not produce adverse effects over a lifetime. The cumulative sum of the HQs for each route is the Hazard Index (HI). Separate calculations are made for children and adults, where children are taken to be aged up to 6 years. The calculations are based solely on noncarcinogenic effects. The use of this methodology for road dust exposure has been developed in detail for the Delta Region, Egypt (Khairy et al. 2011), and more recently for Nanjing, China (Liu et al. 2014). Most of the coefficients used by these and this study are directly

from the EPA guide (USEPA 2002). The primary difference between Khairy and Liu is in the choice of exposure frequency (days/year), using 180 and 350, respectively, reflecting likely exposure of outside workers and permanent residents. This study uses the 350 days/year approach to reflect the exposure of the most vulnerable groups living on or close to the roads.

Only a limited suite of elements: Cr; Ni; Cu; Zn and Pb, have been chosen based on the likely risk when compared to other similar urban environments. The HQs and HIs have been calculated for both children and adults using the minimum, median, mean and the maximum concentrations of each element in each city; these are given in full in Electronic Appendix EA: Table 10. For the purposes of discussion and interpretation, these are further reduced to two scenarios: (1) the greatest risk group—children exposed to the maximum concentration in each city, and (2) the most typical group—adults exposed to the median concentration of each city. These results are summarised in Table 3a, b, where in Table 3a the HI is used to compare the risk between elements and cities and in Table 3b the significance of the ingestion exposure route (HQ_{ing}) is compared to the inhalation (HQ_{inhal}) and dermal (HQ_{derm}) exposure routes.

The only scenario where the Hazard Index exceeds the critical level of 1 is for children exposed to maximum lead concentration in Kolkata (Table 3a, $HI = 2$). In Kolkata, for both adults and children, the order of significance is $Pb > Cr > Cu > Zn = Ni$, whereas in Bengaluru, for children $Cr > Pb > Ni > Cu > Zn$ and for adults $Cr > Pb > Cu > Ni > Zn$. The change in the order of significance of Pb and Cr between cities is the result of the much greater drop in lead concentration between Kolkata and Bengaluru, with only a modest reduction in Cr. For all scenarios, the child HI is between 8.8 and 9.3 times greater than the adult due to the exposure coefficients used. If BAFs had been applied to the HQs, the HIs would have been reduced further. Perhaps of most significance is the effect on the only case where the HI exceeded the critical level: Pb at maximum concentration for children, where this would have reduced this below the calculated value of 2 but would not have reduced it sufficiently to fall below 1. Hazard Index data from Nanjing (E. Liu et al. 2014) are given in Table 3a for comparison. For both children and adults, Nanjing typically has a Hazard Index an order of magnitude lower than Kolkata, with Bengaluru being between the

Table 3 (a) Hazard Indices (HIs) as a comparison of risk between elements and cities; (b) the significance of the ingestion exposure route (HQ_{ing}) compared to the inhalation (HQ_{inhal}) and dermal (HQ_{derm}) exposure routes

	Pb	Cr	Cu	Zn	Ni
Kolkata HI child (max)	2.1E+00	6.3E−01	9.0E−02	5.4E−02	4.9E−02
Bengaluru HI child (max)	6.5E−01	3.6E−01	1.2E−01	5.4E−02	2.7E−02
Nanjing (Liu et al. 2014)					
HI child (max)	8.3E−02	1.7E−01	9.5E−03	2.7E−03	7.4E−03
Kolkata HI adult (median)	5.8E−02	4.8E−02	3.7E−03	1.9E−03	1.2E−03
Bengaluru HI adult (median)	3.6E−02	1.9E−02	2.1E−03	1.5E−03	6.1E−04
Nanjing (Liu et al. 2014)					
HI adult (median)	2.9E−02	4.9E−02	5.3E−03	9.2E−04	2.1E−03
	Pb	Cr	Cu	Zn	Ni
HQ_{ing}/HQ_{inhal} (adult)	6800	65	6800	6800	680
HQ_{ing}/HQ_{inhal} (child)	35,789	341	35,789	35,789	3579
HQ_{ing}/HQ_{derm} (adult)	38	5	75	50	68
HQ_{ing}/HQ_{derm} (child)	54	7	107	71	96

Data for Nanjing (Liu et al. 2014) are given for comparison. The only significant Hazard Index ($HI > 1$) is highlighted in red and is for children, in Kolkata, exposed to the maximum Pb determined in this study

two. The exception is Pb in Kolkata, being 26 times higher than Nanjing. These data suggest that both Kolkata and Bengaluru road dusts provide more of a hazard than those of Nanjing; although, except Pb for children in Kolkata, below the critical level.

The importance of the ingestion route is shown as it dominates the Hazard Quotient; in this case, it is approximately two orders of magnitude larger than dermal exposure and five orders of magnitude greater than inhalation; with such a large difference in HQ, the application of BAFs to the ingestion values would have not substantially changed its importance. Table 3b records the dermal and inhalation HQs relative to the ingestion HQ and demonstrates that this is ubiquitous for Pb, Cu and Zn. The dermal exposure route ratio varies from a factor of approximately 50–100 for Pb, Cu, Zn and Ni but is much more important for Cr at 5–7 (both adults and children). The inhalation route is relatively insignificant ($HQ_{ing}/HQ_{inhal} = 65–35,789$) in all cases, but the most significant scenario is Cr exposure in adults.

Conclusions

This study aimed to evaluate the potential health impacts of road dust in two contrasting cities in India:

Kolkata, a city with a long history of high population density and associated industry, and Bengaluru, a new and rapidly growing city with an emphasis on high-technology industries. Samples were collected from a range of urban sites from high-traffic-density road junctions to urban background in park areas. When considering total elemental concentrations, major geogenic elements demonstrated a difference in base mineralogy between cities; whilst classic anthropogenic elements (Cu, Pb, Zn) were more abundant in Kolkata, presumed to result from its longer industrial and urban heritage. However, techno-anthropogenic elements (Mo and W) were similar between them. When compared to a Road Traffic Index (RTI), a limited number of elements both anthropogenic and geogenic showed an association; Sb, well known to be associated with brake lining wear, was one of these. A limited range of key heavy metal concentrations in the road dust were compared to other cities either in Asia or with similar climatic regimes or development stages reported in the literature. Across these elements, when considering both median and ranges of concentration, both Kolkata and Bengaluru were in the middle of the order. This indicates that both cities although contaminated are not unusual for the large urban environments of megacities. Enrichment factors (EFs) relative to

crustal abundance were calculated, and Kolkata proved to be more contaminated with eight elements at significant enrichment or higher compared to only five in Bengaluru. The high enrichment factors for a number of elements coupled with their low crustal abundances (Cd, Mo, Se, Ag, Sb, Hg and Bi) suggest these would make excellent indicators of anthropogenic activities if sources with anomalous concentrations can be positively identified. Unfortunately, there are currently very little comparative data for these elements in terms of either urban abundance or characterised source material; this may be expected to change in the future, as ICP-MS as an analytical technique with sufficient sensitivity becomes more ubiquitous within environmental chemistry laboratories.

Factor analysis was performed to identify multi-element signatures. When factor scores were compared between city and RTI, for the most important factor 1, Kolkata demonstrated a more contaminated multi-element signature, which became more pronounced with RTI; this was analogous to an undifferentiated urban signal described by others, caused by a plethora of small diffuse sources. Kolkata again was more important in Factor 2 showing a loading associated with fossil fuel burning or possibly steel elements. However, Bengaluru dominated Factor 3 and changed with RTI; this was loaded for elements associated with brake linings; it is proposed that this is observed because contamination from road traffic is observed over a lower long-term industrial background when compared to Kolkata.

Human health assessment started with determining the bio-accessible fraction (BAF) in selected road dust samples from both localities. Key health-related heavy metals were considered (Cr, Ni, Cu, Zn, As and Pb), and in all cases, except As, the stomach phase gave rise to higher bio-accessibility. When considering the median BAF, the descending order of accessibility was $Zn > Pb > Cu > As > Ni > Cr$. No elements showed a relationship between BAF and total metal concentration except Zn for which the relationship was nonlinear. A formal human health risk assessment using the US EPA framework showed that the element of most concern amongst Cr, Ni, Cu, Zn and Pb was Pb in Kolkata, whereas it was Cr for Bengaluru, although Pb was the second most important. In terms of the pathways relative risk; ingestion dominated the hazard quotient, being greater than dermal and much greater

than the inhalation route. The only risk combination (Hazard Index) shown to be significant was lead exposure for children in Kolkata, and this was not found to be high.

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