

# Microzonation, ecological risk and attributes of metals in highway road dust traversing through the Kaziranga National Park, Northeast India: implication for confining metal pollution in the national forest

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**Abstract** Despite the abundant literature on metal contamination through road dust (RD) in urban/suburban and residential/highway regions, the RD of highways traversing through the Kaziranga National Park, NE India, has not been studied and lacks baseline data. The objective of the present study was to ascertain the possibility of highway microzonation based on temporal and spatial variability of metal pollution level and ecological risk. It was found that the RD contains an average content of (1.7–33.5 mg/kg) for Cd, Co, Cu and Pb and (121–574 mg/kg) for

Ni, Zn, Cr and Mn across the highway passing through the national forest attributed by various sources. The study revealed three possible microzones present in the studied highway NH-37 based on spatial trend of metal as well as human interference. An attempt was made to understand the possible source of metals by principal component analysis, and four sources were identified: Three were of vehicular origin, and another was related to road surface and subsurface condition. The use of noise barrier walls as an effective measure to control the translocation of RD from one place to other was recommended to reduce the hostile effects of metal accumulation in the sensitive ecosystem. Thus, the study suggested and necessitated micronizing the system based on human interference level, ecological risk factors, spatial variability of pollutants and traffic pattern for their efficient management and conservation.

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## Introduction

Metals constitute an important group of contaminants, originating from both natural and anthropogenic sources. Metal do not degrade but translocate from one place to other which may impact environment and health (Jankaite and Vasarevicius 2005; Das et al.

2015). Metals can become toxic at higher trophic levels, due to bioaccumulation and magnification in the food chain (Zhao and Zhao (2012; Gogoi et al. 2015); Ayomi et al. 2018). The anthropogenic origin of these metals includes vehicular exhaust particles, tire wear and tear, brake lining wear, weathered street surface particles, power plants, coal combustion, metallurgical industry, auto repair shops, chemical plants and dispersion of construction material (Wang et al. 2009; Khairy et al. 2011; Kumar et al. 2009, 2010, 2013a, b). Hazardous metal species generally accumulate around roadside zones (Akhter 1993; Garcia and Millan 1994; Sutherland et al. 2000; Li et al. 2001; Al-Khashman 2004; Hoque et al. 2007; Ewen et al. 2008; Pagotto 2010; Duong and Lee 2011; Shi et al. 2013). Studies on road dust (RD) are of concern mainly due to two reasons. Firstly, the fine particulate fraction in RD may be being freely inhaled by those traversing the roads and residing within the vicinity of the roads, which might lead to health hazards. Secondly, the seasonal monsoon may transport the RD to the aquatic environment, polluting the water bodies. Metals may become concentrated on the surface sediments of the river bed. This might prove toxic to aquatic life, and, as a worst case scenario, may enter the food chain through contaminated fish, and thus, it has direct impacts on the health of individuals consuming the fish (Allen and Hansen 1996; Chirenje et al. 2006; Christoforidis and Stamatis 2009; Inyang and Bae 2006).

Extensive studies on RD contamination of expressways and other highways of urban regions have been reported in the literature. Swietlik et al. (2015) studied the mobility characteristics and chemical fractionation of metals in the RD retained on noise barrier walls along an expressway in Poland; it was reported that the samples from the noise barrier wall represented a pattern of road traffic density, emission and driving conditions. Lu et al. (2008) and Ram et al. (2014) characterized the RD with metals suspended on plant canopies, and the canopies of the plant were used as a passive metal monitoring system. Jayarathne et al. (2018) investigated the effect of precursor dry weather on adsorption behavior and the transformation characteristics of metals in RD. Aslam et al. (2013) studied metal contamination of RD in Dubai (United Arab Emirates) at different sites with one and more than one traffic signal, and no traffic signal. It was observed that metal contamination had a good correlation with the

no of traffic signal points, signal distance and vehicular attributes.

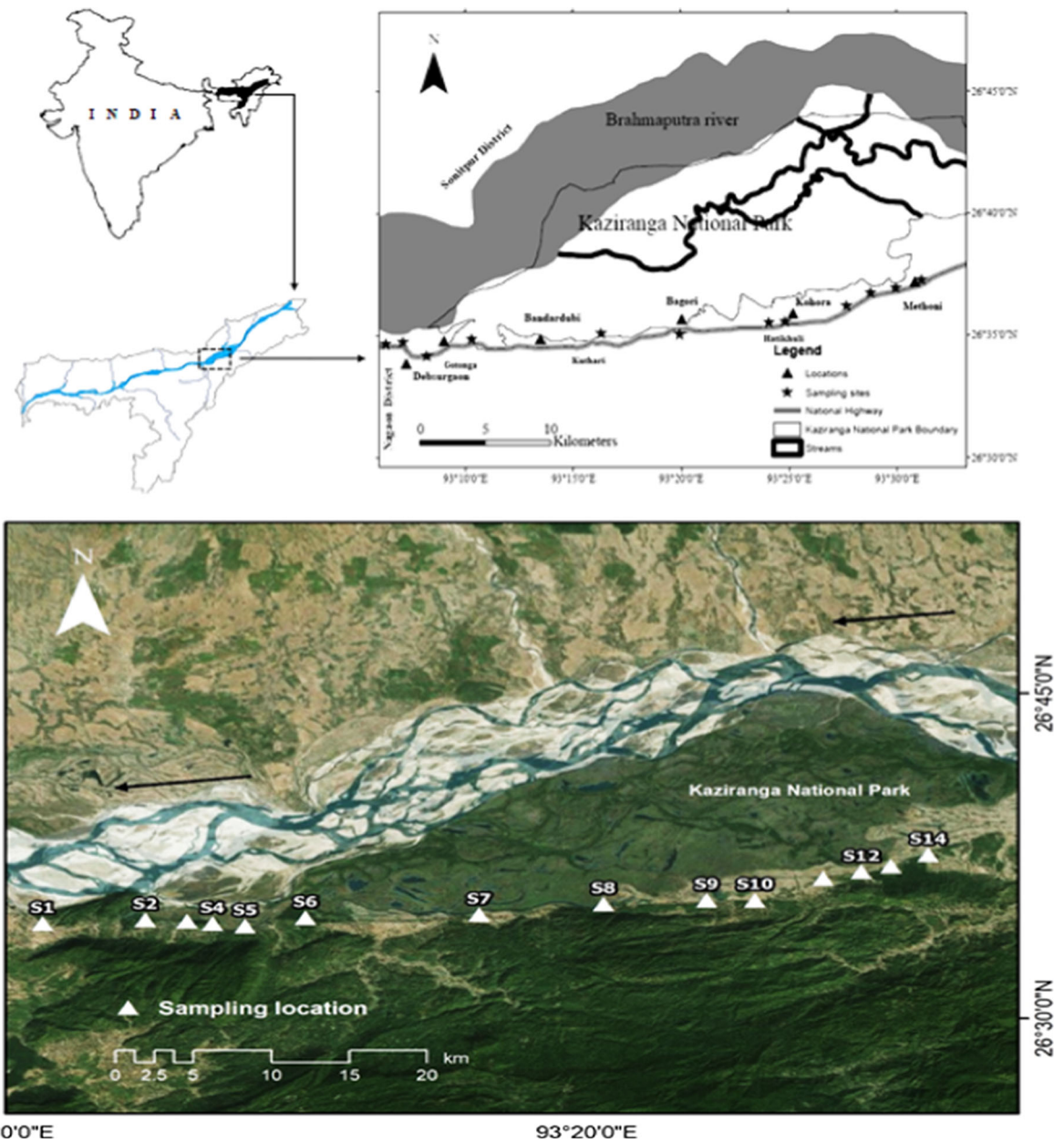
Metal input from highways can be a useful indicator of the level and distribution of contamination in the surface environment (Charlesworth et al. 2003; Ahmed and Ishiga 2006; Al-Khashman 2007; Xiang et al. 2010). Highways that pass through fragile ecosystems like a national forest, wildlife sanctuaries and biosphere reserves add loadings of metals into the environment. This aspect is seldom addressed by researchers. The present study investigates attributes of metals in RD of a national highway (NH-37) passing through Kaziranga National Park (KNP), Assam (India) (UNESCO World Heritage site), during pre- and post-monsoon season. Physical transformations can be seen and felt such as habitat destruction, forest reduction area, frequent flood and animal poaching, and these are reported based on the observation for the entire study period and interview with the local dwellers; however, chemical agents and their effects cannot be predicted until a thorough systematic study has been carried out.

Under the light of above discussion, the present study aims to (a) determine the level of metals, viz. Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn and Cd, in RD of KNP stretch of NH-37; (b) understand the metal distribution and its seasonal and spatial variation in the RD, (c) quantify the metal pollution load carried by RD of NH-37 and its associated potential ecological risk, (d) estimate the metal fractionations and the associated ecological risk, and (e) source apportionment for metals using multivariate statistical techniques, and (f) to propose road dust management plan along the highway traversing through sensitive ecosystem like national forest.

## Materials and method

### Study area

The KNP covers three administrative districts of Assam (Fig. 1). It is bounded by the Brahmaputra river on the north and Karbi-Anglong hills on the south, covering an area approximately 430 km<sup>2</sup>. The region experiences sub-tropical monsoon type climate with an average rainfall of 1320 mm. There are human settlements, agricultural fields, tea gardens and tea processing factories all along the length of the



**Fig. 1** The study area is Kaziranga National Park and the sampling stations are marked on NH-37

highway, beside the park. The NH-37 runs through the park for a stretch of 48 km, and 14 representative locations were chosen within the entire length of the highway for the collection of RD. Sampling locations and prevailing activities are given in Supplementary Table 1. The distance between sampling points was

not uniform in order to sample specific land cover/land use types across the area.

**Sampling**

Four rounds of sampling of RD were carried out, forming a time series, alternately for pre-monsoon and

post-monsoon seasons, during the year 2010–2012. Fifty-six samples of RD were collected during the entire period. The samples were collected on a particular day which was preceded by at least 15 rainless days to avoid rain-washing of RD. Samples were collected by sweeping an area of 0.5–1.0 m<sup>2</sup>, with a brush and dustpan, in order to obtain sufficient volume of sample for analysis. The samples were then transferred to the laboratory in self-sealing polyethylene bags. Collections were made from both sides of the highway and mixed thoroughly to have composite samples to overcome the problem of localized spatial bias. Samples were dried and sieved through a nylon sieve (500 µm) to exclude any extraneous matter such as glass or other plant debris. Care was taken to avoid contamination of samples during sampling, drying and storage.

#### Total metal and sequential extraction

RD sample was acid-digested for metal analysis by microwave aqua regia + HF method, earlier used by Chen and Ma (2001), 1 gm of the sample was transferred into a Teflon bomb, and 4 ml of HF (Hydrogen Fluoride) was added. The mixture was allowed to react for 16 h at room temperature. Twelve milliliters of aqua regia was added and heated in a hot air oven for 8 h. After digestion, 2 gm of boric acid was added to neutralize the excess HF and left until analysis. Samples were filtered, and the filtrate volume was made up to 25 ml with ultrapure water. The metals were quantitatively analyzed by ICP-OES (PerkinElmer/Optima 2100DV). The pH was measured in sample slurries with RD to water ratios of 1:2.5 (Clesceri et al. 1998). The percentage of organic matter of the sample was determined by chromic acid digestion method (Walkey 1947).

Samples from site S2, S5 and S11 representing the eastward, middle and westward reach, respectively, of the NH-37 were analyzed for speciation content. An operationally defined sequential extraction procedure (SEP) originally proposed by Community Bureau of Reference (BCR) and modified by Rauret et al. (1999) was used for sequentially digesting total metal into three fractions, i.e., exchangeable, reducible and oxidizable to represent, respectively, metal associated with carbonates, Fe/Mn oxides, and organic and sulfide content. This three-stage chemical extraction scheme extracts adsorbed metals from the potential

mobile phases in the soil in the order of decreasing mobility. The exchangeable fraction is termed the labile pool (LP) and has the highest mobility, while the sum of an exchangeable fraction, carbonate, reducible and oxidizable fractions is called the potential mobile pool (PMP). The fraction of metal which was retained after the analysis was bound in a crystal structure, hence indicating the residual fraction. Hence, a general idea about the bioavailability and physico-chemical behavior of metals could be made using this approach (Tessier et al. 1979; Benson et al. 2013).

#### Calculation of sorption coefficient ( $K_d$ )

Sorption coefficient ( $K_d$ ) can generally be understood as the ability of a metal contaminant including that of metals to separate out into the water systems from the solid phase. Thus,  $K_d$  characterizes metal mobility potential (Kumar et al. 2009), and the sorption coefficient is the ratio of the amount of metal contaminant in the soil or sediment ( $C_T$ ) to the amount that has been leached out as an exchangeable fraction by the SSE method ( $C_L$ ). This method for ( $K_d$ ) was devised by the US Environmental Protection Agency (USEPA 1999) USA Environmental Protection Agency (1999). ( $K_d$ ) is given by:

$$K_d = \frac{C_T}{C_L} \quad (1)$$

#### Quality control and quality assurance

For each sample, a fresh set of dustpan and brush was used in order to avoid cross-contamination. Standard reference material (SRM 8704) from NIST, USA, was used to assess the accuracy of analytical test results. The recovery percentage for the eight observed metals ranged between 80% and 110%. In order to determine the precision of the analytical procedure, triplicate analyses were performed with random samples exhibiting RSD < 15%, and the reagents used were of analytical grade. Freshly prepared ultrapure water was used for preparation and dilution of the solutions. Reagent blank analyses were also conducted for each new batch of samples to assess the background concentration. All the glassware and plastic vessels were soaked with 10% HNO<sub>3</sub> for 20 h and rinsed with ultrapure water before use.



Statistical analyses

In order to determine the relationship among metals in the RD samples and their possible sources Pearson’s correlation analysis, principle component analysis (PCA) was used. PCA ascertained the possible contributing factors toward the elemental concentration, thereby determining which elements have a common origin in RD. The provenance study was done by applying varimax rotation with Kaiser normalization (Jolliffe 1986).

Calculations for metal indices

Enrichment factor (EF)

Enrichment factors are calculated to evaluate the influence and degree of anthropogenic activities by normalizing the measured metal content in a matrix. EF of an element X (EF<sub>X</sub>) was calculated as follows:

$$EF_x = \frac{|C_{x(\text{sample})}/R_{(\text{sample})}|}{|C_{x(\text{reference})}/R_{(\text{reference})}|} \tag{2}$$

where (C<sub>x</sub>) is the concentration of X in RD and (R) is the concentration of reference element (Fe) in RD.

Fe was chosen as the reference element as it is a geochemical marker owing to its crustal abundance and availability of background concentration data (Zoller et al. 1974; Saeedi et al. 2009). Here, the reference values are background concentrations of metals of the study area (Supplementary Table 2). EF value of less than 5 was considered insignificant because such increased level may arise from differences in the composition of local soil material and reference soil used in EF calculations (Faiz et al. 2009). On the basis of the enrichment factor, contamination was categorized into 5 types. EF < 2 deficiency to minimal enrichment; EF = 2–5 moderate enrichment, however, both of which are considered insignificant; EF = 5–20 significant enrichment; EF = 20–40 very high enrichment; and EF > 40 extremely high enrichments.

Pollution index (PI)

Pollution index (PI) is commonly used to assess environmental quality. PI was calculated as

$$PI = \frac{C_n}{B_n} \tag{3}$$

where (C<sub>n</sub>) refers to the concentration of an element in the RD and (B<sub>n</sub>) the concentration of the element in the background. PI of each element was calculated, and the pollution level of the region was determined on the basis of the classification (Fyfe, 1974; Luo et al. 2012). A PI ≤ 1 signifies low pollution, 1 < PI ≤ 3 moderate pollution and PI > 3 high-level pollution (Hu et al. 2018).

Ecological risk factor

The single index of ecological risk factor (E<sub>r</sub><sup>i</sup>) and the potential ecological risk (RI) as suggested by Hakanson (1980) are as follows:

$$E_r^i = T_r^i \times C_f^i \tag{4}$$

$$C_f^i = \frac{C_n^i}{C_{\text{ref}}} \tag{5}$$

$$RI = \sum E_x^i \tag{6}$$

where C<sub>n</sub><sup>i</sup> is the concentration of metal in the sample, C<sub>ref</sub> denotes the background value of the element, and (T<sub>r</sub><sup>i</sup>) is the toxic response factor of a substance given by Hakanson (1980). Hakanson’s interpretation of E<sub>r</sub><sup>i</sup> and RI values is given in Supplementary Table 3.

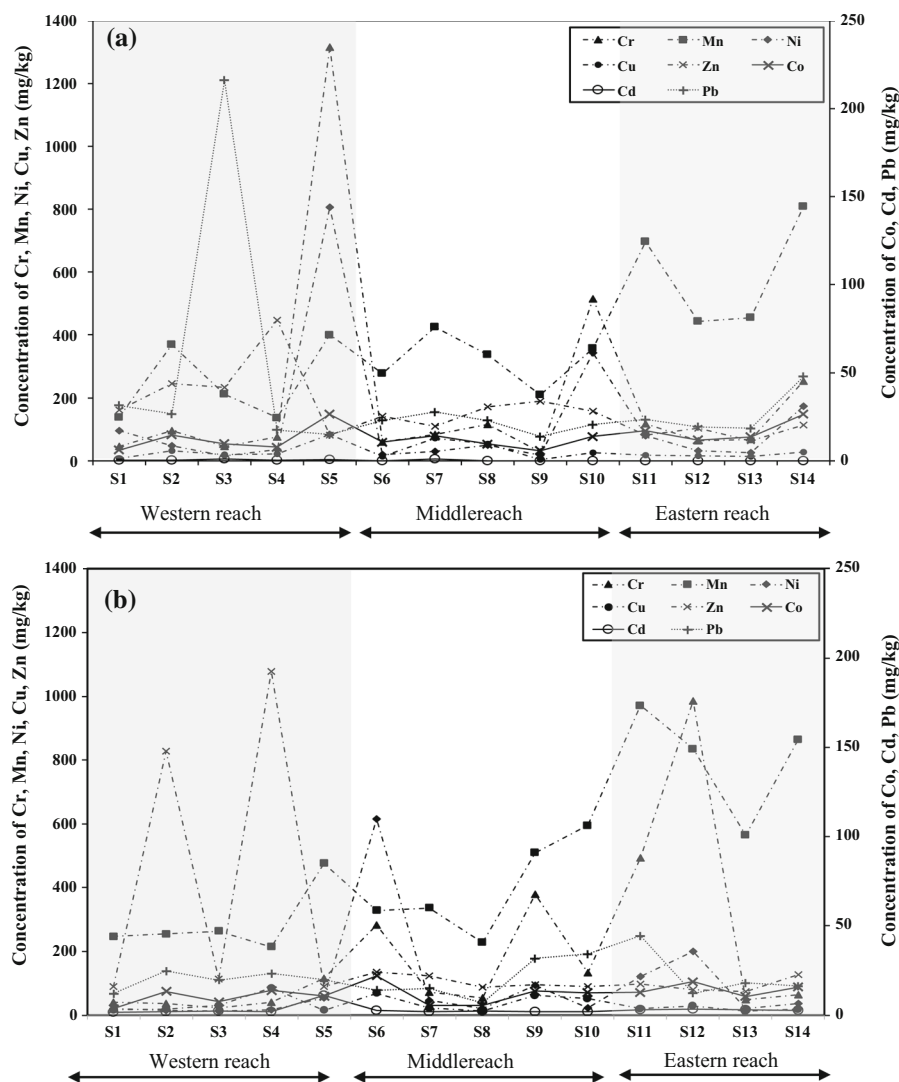
Results and discussion

Trends of metal content

Supplementary Table 2 shows the statistical parameters (minimum, maximum, mean and standard deviation) of metals in RD during pre- and post-monsoon season. The data obtained showed effect of seasonal variation on the concentration of metal across the NH-37. The mean metal concentration was higher for all metals in the post-monsoon samples. This may be because of the change in the no. of vehicles and type of vehicle due to the seasonal variations.

There is a sudden rise of traffic on NH-37 during the post-monsoon season for three principal reasons. First, the end of monsoon brings in dry weather that favors easy movement of goods. During monsoon, which begins in June and ends in September, several regions

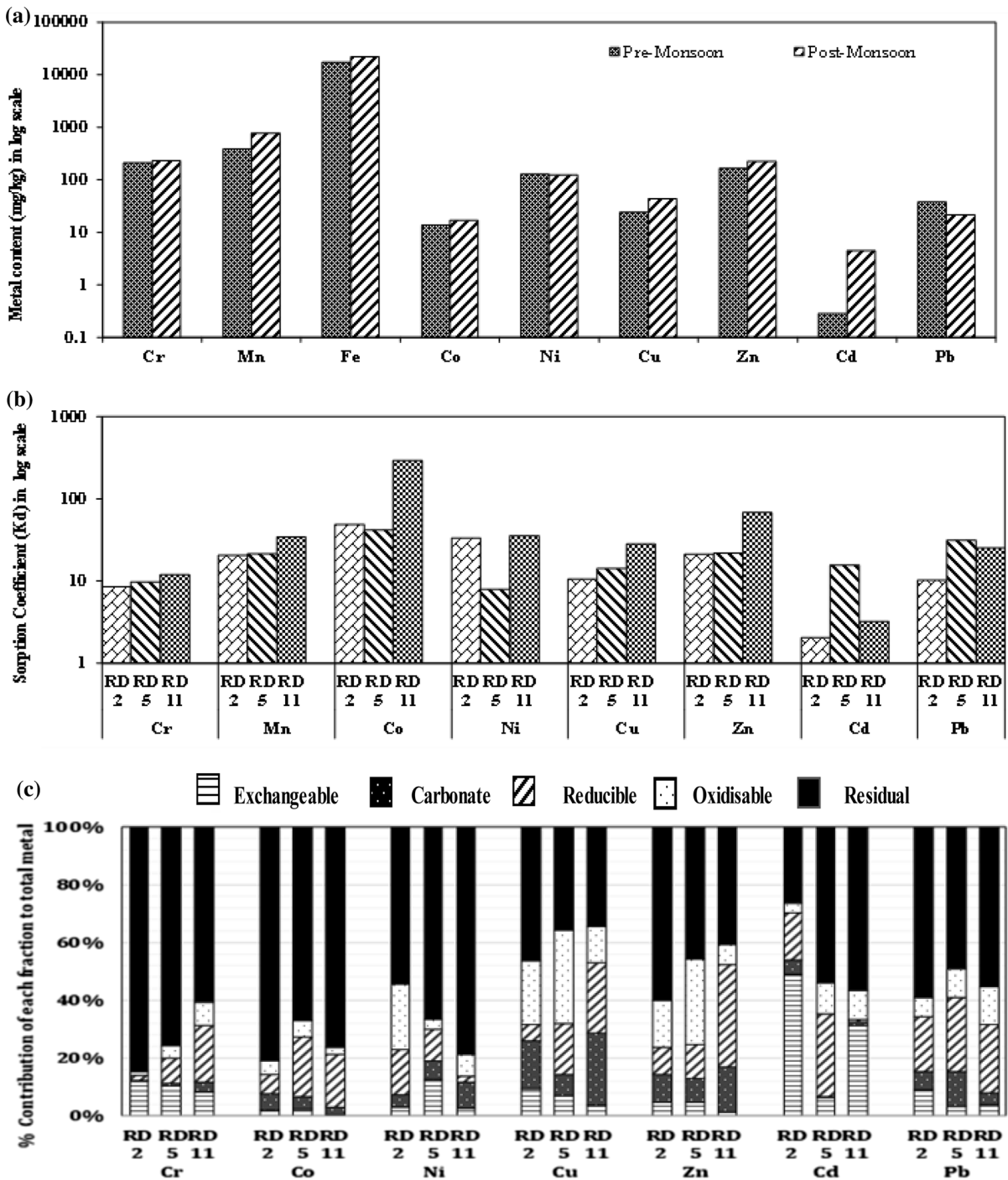
**Fig. 2** Spatial trend of metals along NH-37 during pre-monsoon (a) and post-monsoon seasons (b). The concentration of metal after post-monsoon season was higher at the two extremes of NH-37. Three potential microzones: zone I: western reach (S1–S5), zone II: middle reach (S6–S10) and zone III: eastern reach (S11–S14), were identified based on the metal concentration and spatial trend



of the hill states and parts of Assam remain inaccessible. The demand for food and goods rises tremendously in these areas after the long-wet season. Second, the financial year in India starts on April 1 and ends on March 31 of the following year. Financial planning of government and private business houses always take ‘financial year ending’ into consideration. Therefore, the pace of development activities and volume of supply of food and materials from one corner to another in India depends on cyclical activities related to the financial year. The post-monsoon period is the time when the financial year comes to its second half, and hence, higher economic activity is observed which is marked by a larger volume of supply to meet the higher demand leading to

greater traffic density on major highways. Finally, the reopening of the park during this season attracts more tourists, both national and international, which invite additional traffic load on NH-37 in the stretches of KNP. Hence, a sudden rise in metal concentration during post-monsoon could be explained by the higher traffic density on NH-37 during the same period.

The concentration of metal species was low in the pre-monsoon samples, which could again be attributed to low traffic during this season. The movement of heavy vehicles fall drastically after the ‘financial year end’ (March 31) and also due to occasional rainfall caused by local disturbance, locally called *Bordoisila*. These two factors might explain the lower density of traffic and, therefore, lower levels of metal



**Fig. 3 a** Temporal trend of total metal content in the pre-monsoon and post-monsoon seasons. Except Pb and Ni, all other metal shows high total metal content in post-monsoon, and this trend may be attributed to the bad road condition. **b** Spatial variation of sorption coefficient ( $K_d$ ) along NH-37. Sorption

coefficient for metals at different representative location followed an increasing trend from west (zone I) to east (zone III) except for Pb, Ni and Cd, which shows they are more governed by point source. **c** Metal fractionation of the road dust samples of NH-37 by SSEs procedure

**Table 1** A comparative account of levels of metals of the present study and studies elsewhere

Cities	Sample type	Co	Cr	Cu	Mn	Ni	Pb	Zn	Cd	References
Shanghai	Highway	–	159.3	196.98	–	83.98	294.9	733.8	1.23	Shi et al. (2008)
Kavala (Greece)	Highway	0.2	196	124	–	58	301	272	0.2	Christoforidis and Stamatis (2009)
Baoji (China)	City road	–	–	123	–	49	408	715	–	Lu et al. (2009)
Aviles (Spain)	City road	–	42	183	–	–	514	4829	22.3	Ordonez et al. (2003)
Lancaster (UK)	City road	1.0–14.6	10–91	–	–	15–126	150–15,000	160–3725	–	Harrison (1979)
Islamabad (Pakistan)	Highway	–	–	52	–	23	104	116	5	Faiz et al. (2009)
Istanbul (Turkey)	Highway	–	111	–	–	–	177	245	–	Guney et al. (2010)
Delhi (India)	Highway	–	171.8	128.98	–	98.78	230	420.78	–	Banerjee (2003)
Xian (China)	City road	–	167.3	94.9	687	–	230.5	421.4	–	Yongming et al. (2006)
Yozgat (Turkey)	City road	24.3	32.7	37.7	852	77	69.2	–	3	Soylak (2003)
Xianyang City (China)	City road	–	135.63	132.17	604.36	69.59	77.3	375.38	0.13	Shi and Wang (2013)
Central Assam (India)	Highway	–	–	–	–	50–155	102–495	1.4–1381	–	Hoque et al. (2007)
Tokyo (Japan)	Highway	–	–	442	–	–	44	651	0.51	Kumar et al. (2010, 2013a, b)
KNP ( <i>n</i> = 56), present study	<b>Highway</b>	<b>15.2 ± 14</b>	<b>208.6 ± 323</b>	<b>33.5 ± 39</b>	<b>574.3 ± 797</b>	<b>123.3 ± 200</b>	<b>29.2 ± 37</b>	<b>191.4 ± 230</b>	<b>1.7 ± 2</b>	
Background levels*	<b>Soil</b>	<b>19.39</b>	<b>121.1</b>	<b>43.66</b>	<b>387.13</b>	<b>62.82</b>	<b>13.88</b>	<b>70.42</b>	<b>0.16</b>	
EF		1	2	1	2	3	3	4	14	
PI		0.8	1.7	0.8	1.5	2	2.1	2.7	10.5	

The bold values indicated the mean metal concentrations of road dust obtained from the present study

\*Obtained by analyzing soil samples of natural soil of the study area, 3 km away from the highway



concentrations during pre-monsoon season. However, the concentration of Pb marked a reversal of the above trend; that is, higher level of Pb was found in pre-monsoon samples. We would like to attribute this to change in the composition of vehicles. During pre-monsoon period, there is a surge in the number of lighter vehicles, which run on petrol, whereas the number of heavy vehicles decreases due to a fall in the demand for goods (Devi et al. 2015; Tamuly and Devi 2014).

Though all RD samples were from one single highway stretch having uniform traffic density, an interesting spatial variation of metal concentrations among sampling points was found. The highway stretch was demarcated into three reaches: S1–S5, S6–S10 and S11–S15. Both the initial reach (from the east) and final reach (toward west) connected the urban regions. The middle reach was relatively pristine, with a minimal urban contribution. The spatial trends and temporal trend, from S1 to S14 of metals, are illustrated in Figs. 2 and 3a, respectively. The middle reach showed less magnitude and variability in metal concentration, while the metal concentration in eastward and westward reaches indicated the influence of human proximity. The high and inconsistent values of metals could be attributed to road geometry, the speed of traffic and surface condition of the road. The road geometry has a direct impact on the decelerating, accelerating and climbing actions of a vehicle, hence the different concentrations of emissions by the vehicular engine also, and the road geometry and surface conditions have a definite bearing on vehicular wear and tear, which thus control the metal pollutant load of RD (Pal et al. 2012). There are other low-intensity human activities that prevail on the roadside as illustrated in Supplementary Table 1. These, however, may not have sufficient impression on the metal concentrations except for S5, which is near a vehicle repair workshop, showing higher Co, Cr, Fe and Ni. Other stations did not show such coherence and, therefore, road geometry and surface conditions could be said to have played a greater role in bringing about spatial variation.

Table 1 shows the mean metal concentrations ( $n = 56$ ) of RD of NH-37 in the stretches of KNP together with a comparative account of metals reported for highways and city roads elsewhere. The mean concentrations of Co, Cr, Cu, Mn, Ni, Pb and Zn, particularly Cd, were higher than their background

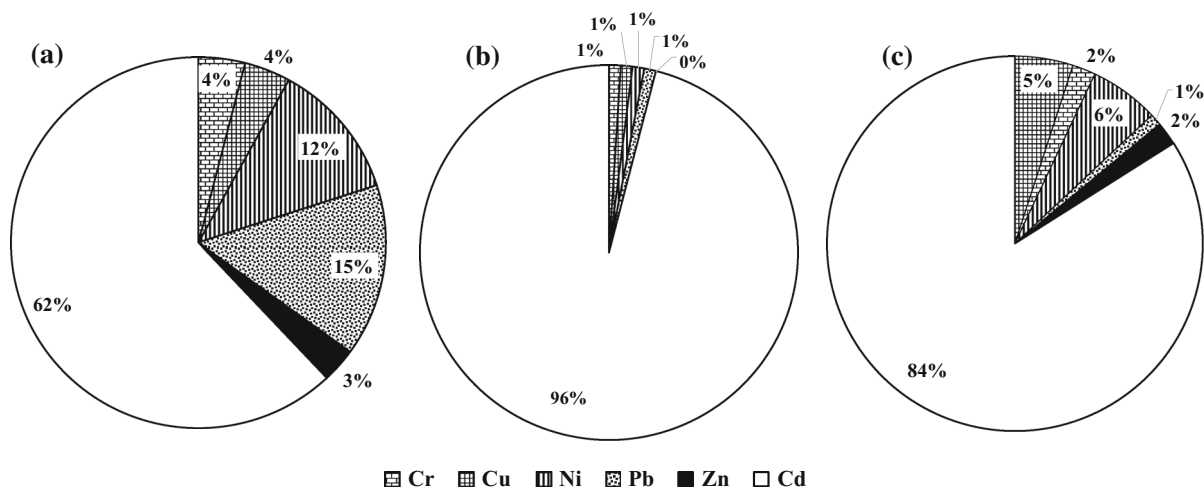
values, suggesting that these metals in road dust of highways in the stretch of KNP were influenced by anthropogenic sources. It was found that Pb ( $29.2 \pm 37$  mg/kg) and Zn ( $191.4 \pm 230$  mg/kg) concentrations in RD of KNP were far below the concentrations reported from highways of China (Shi et al. 2008; Wang et al. 2009), Spain (Ordóñez et al. 2003) and Greece (Christoforidis and Stamatis 2009). The gradual decreasing trend of Pb concentration in RD has been observed due to phasing out of leaded gasoline (De Miguel et al. 1997; Yongming et al. 2006). Pb level of 230 mg/kg reported from Delhi a decade back by Banerjee (2003) was quite high in comparison with the finding of the present study. As mentioned earlier, due to lack of past studies from the region, the extent of reduction of Pb in the study area after phasing out of gasoline Pb could not be ascertained. However, a high level of Pb in RD from KNP is due to higher vehicular strength on NH-37. However, Ni, Cr and Cd concentrations of the present study were on the higher side compared to the studies carried out in Pakistan (Faiz et al. 2009), China (Shi and Wang 2013), UK (Harrison et al. 1981) and Turkey (Güney et al. 2010; Soylak et al. 2003).

Overall, the metal content in the stretch of this NH is lower than many other highways owing to less traffic density as well as pristine less metal pollution sources. Thus, in order to ascertain the pollution level, background soil pollution level has to be taken into consideration. Therefore, the next section will estimate the pollution level based on different metal indices.

#### Metal pollution level through various indices

##### *Enrichment factor (EF)*

Table 1 shows the mean EF values. EFs of Co, Mn and Cr were slightly less than 2, and those of Cr, Cd, Ni, Pb and Zn were marked by moderate increased level indicating that these in RD could originate from both anthropogenic and natural sources. The increased level of Zn could be attributed to wear and tear rate of tires (Alloway 1990). Some other sources could be mechanical abrasion of brass alloy, oil leak sumps and cylinder head gaskets of the vehicle in origin and zinc salt containing herbicides in the nearby tea gardens (Amato et al. 2009; Carlosena et al. 1998; Jiries 2003). However, the effect of pesticide spraying in the local



**Fig. 4** Percentage contribution to mean ecological risk of the metals during (a) pre-monsoon (b) post-monsoon (c) for the entire study period. The ecological risk posed by Cd was highest

in both the seasons. There was an abrupt increase from pre- to post-monsoon season which might be due to change in vehicular strength and density

tea gardens on metal accumulation in RD needs detailed further studies. There is a significant accumulation of Cd, as indicated by the high EF. The degree of enrichment of the metals was found in order of  $Cd > Zn > Pb > Ni > Mn > Cu > Cr > Co > Mn$ . Metals on the left are more from anthropogenic activities and, more we go to the right, are more likely to be from natural sources.

#### Pollution index (PI)

Table 1 shows the mean pollution index (PIs) of metals. Cd had the highest pollution level ( $PI = 10.5$ ), while Cu, Co and Mn had caused the least effect on environmental quality. Pb, Ni and Cr fell into the moderate pollution group. However, 14% of samples showed high pollution with respect to Ni. Seventy-nine percent of the samples showed a PI for lead within  $1 < PI \leq 3$ , that is, moderate pollution; 7% of the samples lie in the third quartile, that is,  $PI > 3$ , which is classified as high level of pollution. Zn also showed moderate to high PI values; 72% of the samples showed a PI value within  $1 < PI \leq 3$ , that is, moderate pollution level; 14% of the sampling stations lie in high pollution group, and the highest PI value was found to be 9.34. Hence, there is a marked spatial control over the polluting capacity of individual metals. This can be attributed to their variable concentration, along with the sampling stations.

#### Ecological risk

Supplementary Table 3 shows the minimum, maximum and the mean  $E_r^i$  and RI of metals in the present study. Mean  $E_r^i$  of Cd (835) indicates that Cd poses a very high ecological risk to the biota of KNP. Maximum  $E_r^i$  values of Cu, Ni and Cr fall in the category of considerable risk. Cumulative mean RI (952) is indicative of posing a very high ecological risk, and hence, it is a major threat to biodiversity. The percentage contributions of maximum ecological risk posed by various RD metals of NH-37 are illustrated in Fig. 4. Cd accounted for the maximum risk as compared to other metals; similar results were also reported by Shi et al. (2010) for a suburban area and Zhang et al. (2013) for street dust. This could be the result of anthropogenic input of Cd from various human activities around NH-37. Wei et al. (2009) attributed anthropogenic inputs of Cd into road dust to commercial activities, industrial activities, application of organic manures and phosphatic fertilizer in the parks. Wei and Yang (2010) reviewed a few metal contaminations-related studies in several cities from China over the past 10 years and found that agricultural soils are also significantly influenced by Cd, Hg and Pb derived from anthropogenic activities. There are tea gardens and agricultural areas which use organic manure and phosphate fertilizers in KNP. The

**Table 2** Factor loadings for varimax-rotated PCA of metal data in road side deposited sediments and corresponding meaning of loading through predicting source apportionment

	PC1	PC2	PC3	PC4	Communalities
Co	<b>0.836</b>	<b>0.481</b>	0.075	0.023	0.936
Cr	0.057	<b>0.870</b>	0.252	− 0.086	0.831
Cu	<b>0.724</b>	<b>0.579</b>	− 0.033	− 0.004	0.860
Fe	0.304	0.221	<b>0.759</b>	− 0.210	0.762
Mn	<b>0.891</b>	0.263	0.078	− 0.050	0.872
Ni	0.233	<b>0.863</b>	0.121	0.045	0.817
Pb	<b>0.743</b>	− 0.237	0.027	− 0.022	0.609
Zn	− 0.059	0.053	− 0.155	<b>0.871</b>	0.788
Cd	− 0.156	0.272	<b>0.713</b>	− 0.072	0.612
OC	0.083	− 0.236	<b>0.547</b>	<b>0.477</b>	0.589
Eigenvalues	3.9	1.598	1.155	1.022	
% Variance	38.99	15.98	11.55	10.22	
% Cumulative	38.99	54.98	66.53	76.75	
Source apportionment	Vehicular exhaust particles	Wear and tear of vehicular parts	Organic manure and fertilizers	Wear and tear of rubbers	

(Observations = 56; variables = 10)

Bold values showed higher loadings of particular metals compared to other metals

Cd could, therefore, be a product of agriculture and tea plantation activities.

In summary, different indices do indicate the enrichment, factor and risk threats for different metals; however, all indices presented in this section are based on total metal content. It is a well-established fact that total metal is not good indicator of actual bioavailability, metal toxicity and environmental safety. Therefore, we decided to carry out speciation study to be presented in the next section.

#### Metal speciation using selective sequential extraction

Figure 3c represents the inherent spatial variability in speciation for individual metals. Three main spatial trends could be identified. For Cu, Cr and Zn, the exchangeable fraction decreased from east to west, while the PMP increased. Ni and Cd exhibited a decreasing trend in PMP from east to west, indicating anthropogenic control, on both sides of NH-37 while Co and Pb concentration was highest in the middle reach. Co and Pb are released from the particulates present in the vehicular exhaust; hence, the ecology of the middle reach is most affected by the particulates from vehicular exhaust.

It can be seen from Fig. 3c that all the metals contain more than 40% residual fraction which is not mobile; hence, the toxicity of all metals was effectively reduced by the same ratio. Cu, Zn and Cd in S2 contained the least residual metal fraction (< 50%) and hence posed the most ecological risk. Cr, Co and Ni showed a low level of labile fraction (~ 20%, 30% and 35%), and hence, the ecological risk could be inferred to be comparatively less (Fig. 3c). Pb poses a moderate environment risk, hence corroborating the results of ecological risk assessment using the pollution index and ecological risk factor (Table 1). Their chances of plant uptake were highest, and the species could easily transform into toxic metabolites in vivo or during transportation in the ecological system.

Metal fractions not only allow to estimate toxicity but also can help to quantify the sorption/distribution coefficient and thus help to understand groundwater pollution vulnerability from road dust. Therefore, in the next section we present the sorption coefficient estimation and variations.

#### Spatial variation of sorption coefficient along NH-37

Figure 3b represents the spatial variability in the sorption coefficient ( $K_d$ ), indicating the differences in

type and magnitude of polluting sources. The ( $K_d$ ) value found decreased from east to west, i.e., S11–S5–S1. It was observed that for Cr, Mn, Cu and Zn the  $K_d$  values were maximum at S11 > S5 > S2. This indicates that the source of these metals is from the urbanized regions on the east end of NH-37, and this can be well observed from sampling feature as shown in Supplementary Table 1. The value of ( $K_d$ ) was < 50 for all the metals, and for sites except Co (Site S11), the ( $K_d$ ) was almost six times higher. This again is a further indication of the provenance of Co from urbanized areas, as it can be seen that S11 is occupied by tea garden, dwelling houses and agricultural fields. The anthropogenic action and the fertilizers being used in the tea garden and agricultural field might have resulted in an abrupt increase in  $K_d$ . For Co, Ni, Cd and Pb, the sorption coefficient was almost highest in the middle reach except for Ni, indicating that vehicles are the sole contributor of these metals to the environment. The labile fraction of Co is negligible, and hence, high ( $K_d$ ) further indicates a high fraction of non-labile forms. Therefore, on the basis of ( $K_d$ ) it is possible to quantify the retention order or potential mobility of different metals in each sample.

#### Statistical source apportionments

So far we have understood the level, distribution, variation and risks of metal pollution in the road dust of highway passing through this ecological sensitive ecosystem. The next logical step is to understand the source of pollution, for which both bivariate correlation and multivariate principle component analyses were carried out.

#### Correlation-based source estimation

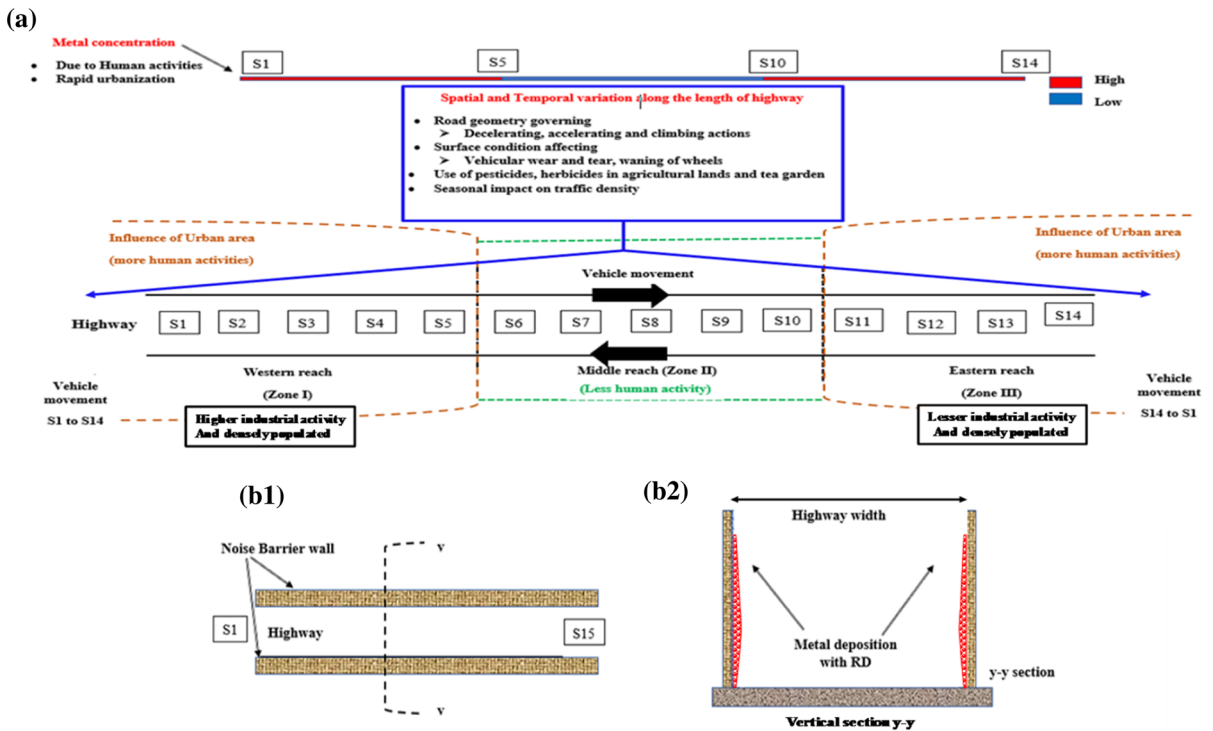
Supplementary Table 4 shows the Pearson's correlations between metal pairs to find interrelationship among metal pairs. Pairs showing significant correlation would mean similar chemistry or origin. The metals like Co and Cu ( $r = 0.87$ ,  $p < 0.01$ ), Co and Mn ( $r = 0.94$ ,  $p < 0.01$ ), Co and Ni ( $r = 0.58$ ,  $p < 0.05$ ), Cr and Co ( $r = 0.43$ ,  $p < 0.05$ ), Pb and Co ( $r = 0.40$ ,  $p < 0.05$ ), Pb and Mn ( $r = 0.43$ ,  $p < 0.05$ ), and Ni and Cr ( $r = 0.80$ ,  $p < 0.01$ ) showed some relationships. The high correlation between Ni and Cr can be explained by their similar origin. Both these metals are used for coating of several parts of

vehicles and, therefore, it is likely that the metals come from vehicular wear and tear. Christoforidis and Stamatis (2009) and Lu et al. (2009) also reported a high correlation between Ni and Cr and attributed them to have come from anthropogenic factors. The relationship of Pb with other metals would mean emission from gasoline-driven vehicles. Mn also showed high correlation with Cu ( $r = 0.83$ ,  $p < 0.01$ ) and Co ( $r = 0.94$ ,  $p < 0.01$ ). Mn is often regarded to have been emitted from diesel exhausts and so, Co and Cu together with Mn could be attributed to diesel exhaust. Positive and significant correlation between Cd and Zn may indicate a common natural source. The negative correlation between Cd and Zn ( $r = -0.02$ ) obtained in this study could mean that there has been sufficient anthropogenic contribution.

#### Principal component analysis (PCA)

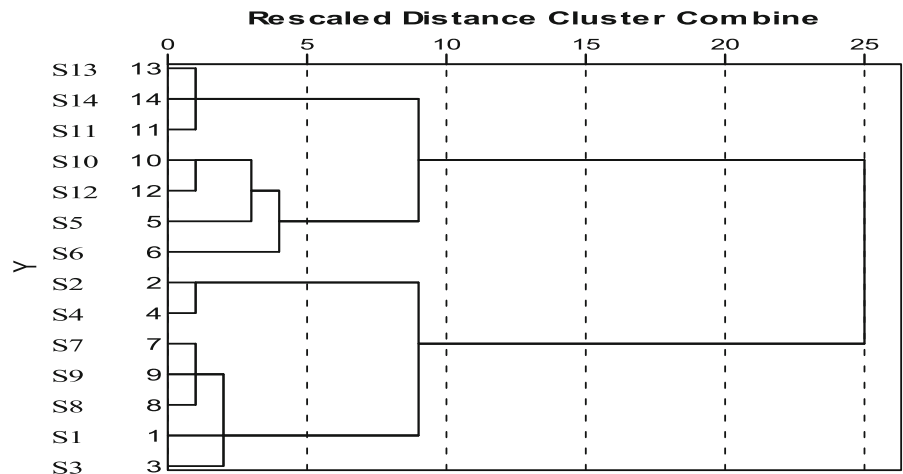
Table 2 summarizes the result of PCA. PCA is often applied to a large set of variables in order to reduce dimensionality and to extract a smaller number of independent 'latent' variables, called principal components (PCs). The analysis provides relationship among measured chemical variables and their multivariate patterns based upon eigenvectors of a variable that offers significant insight. There are several criteria to identify the number of PC to be retained in order to understand a data structure (Jackson 1991; Lin et al. 2002; Loska et al. 2004). In the present context, 4 eigenvalues > 1 were considered that explained ~ 78% of the variance, which then was 'cleaned up' by varimax rotation.

PC1 explained 39% of the variance and showed high loading for Co, Cu, Mn and Pb. Mn is a plausible contribution from diesel exhaust, and Pb and Mn from crustal; therefore, PC1 could be attributed to contributions from mixed sources-exhausts particles from vehicle, both gasoline and diesel, and crustal input. PC2 explained 16% of the variance and is mainly associated with very high positive loadings of Cr, Cu and Ni. This group could be attributed to wear and tear of vehicular parts. As such, Ni and Cr are important coating metals of vehicular mechanical parts, which due to wear and tear are released into the environment. Many vehicular nonmetallic parts are Cr-plated or Ni-plated. There could be input from wear and tear of



**Fig. 5** A schematic depiction of **a** Microzonation of NH-37 into zones 1, 2 and 3 based on spatial trend, traffic pattern and human interference. Proposed solution noise barrier wall: **b1** is plan across the length of the highway and **b2** is the cross section of the wall

**Fig. 6** A dendrogram depiction of similarity and dissimilarity among the locations based on road dust characteristics and metal loading



these parts and could also contribute to the total metal content.

PC3 showed higher loadings for Cd and OC, which could be attributed to organic manure and fertilizer application in tea gardens and agricultural fields. Cd in road dust has been attributed to commercial activities, industrial activities, application of organic manures

and phosphate fertilizer by previous workers (Wei et al. 2009). PC4 showed high loading for Zn and OC, which could be attributed to the waning of wheel rubber. Tire wear and tear have been reported to significantly contribute to the Zn load of RD (Councill et al. 2004). Vehicle tire wear and tear release lots of particulates that are organic and are deposited on the

roadside. Thus, the association of Zn and OC has been well exhibited by the PCA.

Overall, out of the four sources of metals which were identified by PCA, only one (PC1) belongs to exhaust particles from vehicles. The non-exhaust contribution was found to be quite significant in the study. Earlier studies also suggested that non-exhaust sources constitute a substantial amount of vehicular-driven particle emission (Harrison et al. 2001; Ketzal et al. 2007) and that brake lining, tire and road surface wear and tear significantly contribute to the total metal (Thorpe and Harrison 2008).

### Environmental implications of the study

In our final approach, we decided to come up with the microzonation of the entire stretch of KNP for better management of the road dust. Firstly, we micronized the entire stretch based on human interference level, ecological risk factors, spatial variability of metals and traffic pattern into three zones. Figure 5a shows the microzonation of NH-37 across the stretch of KNP, based on the metal contamination three microzones which were identified: zone I (western reach), zone II (middle reach) and zone III (eastern reach). The contamination of RD of NH-37 which stretches along KNP is alarming. The metals being contributed by different agents and source might affect the ecosystem and the local dwellers across the roadside. The toxic effect of the metals can further increase, if it enters into the food chain via direct and indirect ways. Though there are different techniques to control the mobile behavior of metals, some prominently used method is by using various kinds of organic and inorganic sorbents (Balintova et al. 2016). In the present study, the use of noise barrier wall along the stretches of highway in KNP is proposed. The noise barrier will curb the mobility of RD with metals but keeping corridors open and unaffected for the ease of wildlife movement.

Figure 5b shows a schematic representation of the proposed remedial measure. Swietlik et al. (2015) carried out fractionation study and assessed the mobility characteristics of metal with RD deposited on the noise barriers wall along the express way. With time due to an increase in the no. of vehicles passing through NH-37 and other environment agents. The dust particles, particulate matters from vehicle exhaust

may get deposited on the surface of the wall, and the translocation of metal to the ambient sensitive ecosystem can be avoided. Also, the noise barrier wall acts as confining system as well as a separator, thereby protecting the wildlife and ecosystem from the potential ill effects of metals. Also, the noise barrier will reduce the noise pollution and migration of wildlife into open space preventing casualties. Apart from noise barrier, another alternative will be a proper management of the road system such as improving the surface condition.

Finally, in order to support our zonation, we did carry out the cluster analyses to find the similarity and dissimilarity between locations (Fig. 6). We did find a grouping of locations where S11, S13, S14 of zone III, S1 and S3 of zone I and S7, S8, S9 of zone II formed really strong close clustering. Although we must not expect that results based on chemical pollution loading and based on common sense zonation should exactly coincide, the both results showed encouraging similarity and thus necessitate the human interference level, ecological risk factors, spatial variability of pollutants and traffic pattern-based microzonation of the highways for their efficient management and conservation.

### Conclusion

The study effectively illustrated the spatial as well as seasonal variation of RD metal contamination of NH-37 that stretches through ecologically sensitive zone of KNP. The highway stretch was demarcated into three microzones (I, II and III) based on the site locations, sources and level of contamination and pollution levels (enrichment factor, pollution index and ecological risk). It was observed that the RD contamination of western (zone I) and eastern (zone II) reaches was higher compared to the middle reach (zone III) due to significant anthropogenic activities from urban areas and other sources. Explicit temporal and spatial trends of metals were observed, which could be attributed to varying vehicular strength and road conditions, respectively. Except Pb, the abrupt increase in metal concentration from pre-monsoon to post-monsoon season was attributed to change in vehicular strength and density. The poor road condition could be the source of high Pb concentration in RD even in the post-monsoon season. Also Ni, Cr and Cd showed



much higher concentrations compared to results seen in studies from elsewhere in the world. However, levels of Pb and Zn were found to be far below the levels reported by several studies. High levels of Cd have implications on the ecology, which is clear from the calculated levels of PIs and ecological risks (PI = 10.5; EF = 14). Further, the SSE procedure revealed that Cd, Cu and Pb were most labile. Three different trends were observed and followed by metals in a cluster, i.e., (1) Cr, Cu & Zn (2) Ni & Cd and (3) Pb & Co in SSE procedure. Sorption coefficient for metals at different representative location followed an increasing trend from west (zone I) to east (zone III) except for Pb, Ni and Cd, which shows they are more governed by point source. Thus, the high bioavailability of these metals makes them most harmful to the ecosystem of the KNP. Cobalt has the highest retention capacity. Four sources of metals were identified based on the PCA analysis, of which three were of vehicular origin, viz. particulate exhaust released from vehicles, wear and tear of metallic parts and waning wheel rubbers due to abrasion.

The study recommends the use of noise barrier wall as a remedial measure to curb the problem of mobility of metal to the sensitive ecosystem and necessitates microzonation of the highways for their efficient management and conservation based on the human interference level, ecological risk factors, spatial variability of pollutants and traffic pattern. It also emphasizes on good road and vehicle condition that can minimize metal pollution to a great extent. However, more studies are required on other forms of toxins released from the highways like the polycyclic aromatic hydrocarbons (PAHs) and volatile organics to ascertain the cumulative impact on the ecology of the park and also to carry out an effective microzonation study.

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