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ORIGINAL PAPER

Evaluation of heavy metal and total petroleum hydrocarbon contamination of roadside surface soil

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Abstract The present study was conducted along three major highways namely State Highway (SH49), National Highway (NH66 and 45A) connecting Puducherry (India) for assessing heavy metals and total petroleum hydrocarbon contamination from surface soils in close proximity at a depth of 0–15 cm into automobile repair workshops and agricultural fields located beside the highways. Contamination levels of copper, lead, zinc, manganese, cadmium and chromium were assessed in the surface sediments soil on the basis of geoaccumulation index, contamination factors and spatial variability. The results revealed that sampling sites in the proximity to automobile workshops were moderately to considerably pollution impacted as compared to soil from agricultural fields along highways suggesting a direct influence of anthropogenic activities on levels of contamination. The concentration of heavy metals in the surface soil of automobile workshops close to NH66 ranged between 143.07 and 319.28 mg kg^{-1} copper; 68.72–396.41 mg kg^{-1} lead; 162.42–284.91 mg kg^{-1} zinc; 212.72–401.33 mg kg^{-1} manganese; 0.12–15.41 mg kg^{-1} chromium; and 0.73–1.06 mg kg^{-1} cadmium in dry soil. However, in agricultural fields, the concentrations varied between 33.68 and 66.62 mg kg^{-1} copper; 27.22–73.66 mg kg^{-1} lead; 26.24–75.59 mg kg^{-1} zinc; 137.88–242.07 mg kg^{-1} manganese; 0–0.21 mg kg^{-1} chromium; and 1.04–1.58 mg kg^{-1} cadmium. Total petroleum hydrocarbon concentration near automobile workshops ranged between 90.72 and 121.79 mg kg^{-1} in contrast to 44.94–83.4 mg kg^{-1} in agricultural fields. Total petroleum hydrocarbon concentration indicated strong positive correlation with zinc ($r = 0.811$), copper ($r = 0.761$); lead ($r = 0.642$), Mn ($r = 0.571$), chromium ($r = 0.530$) and strong negative correlation with cadmium ($r = -0.765$) at 0.05 significance level. Pearson correlation indicated a strong association of total petroleum hydrocarbon with copper, lead and zinc suggesting that the metal contaminants from roadside surface soil had emanated from a common source.

Keywords Agricultural fields · Automobile workshops · Contamination factor · Geoaccumulation index · Total petroleum hydrocarbon · Transportation

Introduction

As transportation systems become more integrated into the human civilization, their impacts on air, water and soil are inevitable. Some countries in Europe restrict the usage of transportational modes considering their environmental effects (Tuzkaya 2009). In the rural areas, roads usually pass through heavily fertilized crop fields (Ahmad and Erum 2010). Long-term accumulation of heavy metals from road traffic effects soil biota and food. Pb released from emissions gets translocated from the roots to the shoot and leaves in plants (Christiana and Samuel 2013). Urban soils are marked with elevated levels of heavy metals due to extensive anthropogenic activities related to vehicular emissions and traffic, industrial and urban waste, mining, smelting and manufacturing by metal industries, wear and tear of tires, building materials and road activities, fertilizers and agricultural run-off (Lu et al. 2010; Mahanta and Bhattacharyya 2011; Malik et al. 2010; Montoneri et al. 2014). Heavy metal accumulation in soil, their origin and

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interactions with soil properties act as bioindicators in many environmental-monitoring studies (Qishlaqi and Moore 2007).

Automobile repair workshops release waste products such as engine oil, transmission oil, brake fluid, damaged tyres, battery electrolytes, wire carbide, spent batteries and cells into their surrounding areas. Their improper disposal leads to the discharge of toxic emissions during abrasion and contributes to metal contamination of the areas around the auto-repair workshops (Aruleba and Ajayi 2012). Heavy metals pose higher risk for living organisms rather than metals present in higher concentration (Farombi et al. 2013). Scrap batteries and solder, waste engine oil, brake fluid and other fluid generated by their activities in the workshop are not properly disposed off. They are usually released on to the surrounding soil. Since the activity of artisans in auto-repair workshops is one of the major routes for entry of heavy metals into the environment to cause contamination of soil and drinking wells and crops, monitoring the available pools of metals in contaminated soils becomes relevant. Accumulation of heavy metals in the soil possibly controls the soil microbial functions triggering toxicity and contamination of the food chain. Lateral migration of heavy metals towards drinking water wells dug in these reclaimed auto-repair workshop areas causes discharge of heavy metals via soil into wells thereby exposing the residents to unsafe water for drinking and other domestic usage (Ipeaiyeda and Dawodu 2008). The term total petroleum hydrocarbons (TPH) is used to describe a large group of chemical compounds that emanate from crude oil. There are so many different chemicals in crude oil and in other petroleum products, which is not practical to measure separately. Scientists divide TPH into groups of petroleum hydrocarbons that act alike in soil. These groups are called petroleum hydrocarbon fractions. Each fraction contains many individual chemicals (http://www.atsdr.cdc.gov/toxfaqs/tf.asp?id=423&tid=75).

The study of spatial distribution of heavy metals (HM) in soil is necessary in assessment and mapping of soil heavy metals to assess their effects on soil and to define contamination extent for mitigating soil degradation and increasing crop production. The geostatistical method of spatial interpolation is termed "kriging". Because of its unbiased character and advantage in spatial interpolation, kriging has been widely applied in many scientific disciplines. In particular, in soil science, kriging has become an important interpolation method to estimate the spatial distribution of contaminants (Omran and Razek 2012).

The purpose of the present work was to assess the heavy metal contamination in surface soil located adjacent to highways connecting Puducherry, India. Soil samples were analysed for contamination with heavy metals (Pb, Zn, Cu, Mn, Cd and Cr) and for total petroleum hydrocarbons. Metal contamination status was assessed using pollution indices namely geoaccumulation index and contamination factor. Study period was from April 2010 to September 2011.

Materials and methods

Site description

Soil exposed to petroleum hydrocarbon and heavy metal contamination due to traffic and transportation was collected from ten sites near automobile repair workshops from Gorimedu area of National Highway, (NH66) of Puducherry, India. Sampling site was divided into two categories, namely locations adjacent to automobile workshops that include sampling sites 1–10 designated as S1, S2, S3, S4, S5, S6, S7, S8, S9 and S10, and soil from agricultural fields in and around Pondicherry that includes sampling sites 11–16 that are represented by S11, S12, S13, S14, S15 and S16. First two agricultural soil samples were collected from sugarcane fields near industrial area SIPCOT, Cuddalore (National Highway, NH45A), while the remaining four samples were collected from sugarcane, paddy fields and coconut plantation in Marakanam area of Pondicherry beside East Coast Road from Chennai to Puducherry (State Highway, SH49). SIPCOT industrial estates were established in 1982, and the area is located 8 km from Cuddalore on the seaward side of the Cuddalore to Chidambaram East Coast Road (NH45A) Tamil Nadu. The area is marked with high salinity due to discharge from domestic and industrial effluents. Some of closed industries of the area have continuously dumped scrap and chemicals like sulphuric acid, sulphur, lime, pesticides and other raw materials, which were the chief sources of subsurface contamination (Sankaran et al. 2012). Pondicherry University ground soil was taken as relatively uncontaminated control (Fig. 1).

Random collection of samples from each site was done on monthly basis from different locations and at two different depths (0–15 cm) from the soil surface during the period April 2010 to September 2011. Samples were then transported to the laboratory in plastic bags and stored in the refrigerator until further analysis.

Extraction and analysis of heavy metals

To determine the total heavy metal content in soils, air-dried 0.5 g of dry soil was sieved through 250-µm nylon sieve (Addo et al. 2012) and digested in aquaregia using microwave-assisted digestor (Anton Paar, Austria) in accordance with the USEPA method SW 3051 (USEPA 2000).

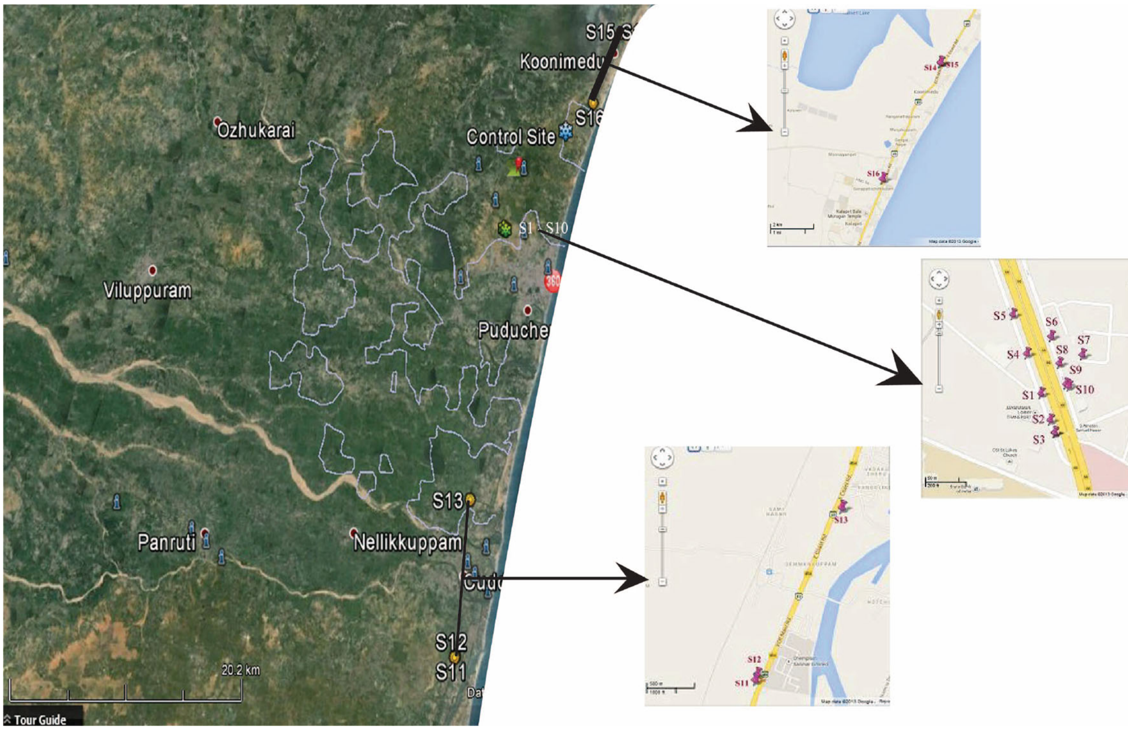

Fig. 1 Location map of the surface soil for sampling sites

Quality assurance

Metal concentrations in the final solution were determined using Atomic Absorption Spectrometer (GBC Scientific Equipment, Australia) that consists of a hollow cathode lamp, slit width of 0.7 nm and an acetylene flame. Zn was analysed at a wavelength of 213.86 nm, Mn at 232.00 nm, Cd at 228.80 nm, Pb at 283.31 nm and Cu at 324.75 nm according to the procedure described in the operation manual (Usha and Vikram Reddy 2012). Standard stock solutions for all the elements suitably diluted following the procedures (APHA 2005). Double-distilled water and analytical grade reagents were used throughout this work. Reagent blank was used to minimize impurity of metal of interest.

Extraction of total petroleum hydrocarbons

Total petroleum hydrocarbon (TPH) was measured with an InfraCal cuvvette holder model Total organic Grease (TOG)/TPH Analyzer according to the USEPA Methods 418.1 that uses S-316 as extraction solvent in replacement to Freon (ASTM 2004). The common method for estimating TPH in soil was USEPA Methods 418.1(Schwartz et al. 2012). This is quick and easy field and laboratory analysis method for determining TPH and oil and grease concentration levels in soil and water. Total petroleum hydrocarbons were calculated from the sample using the formula (USEPA 1978):

$$\text{Concentration of TPHs} = \frac{R * D}{V}$$

where R = mg of petroleum hydrocarbons as determined from the calibration plot; D = extract dilution factor; V = volume of sample, in litres.

Geoaccumulation index (Igeo)

Geoaccumulation index (Igeo) was used to assess the contaminant concentration of sediments and soils (Nava-Martinez et al. 2012).

Geoaccumulation index (Igeo) was calculated using the formula:

$$\text{Igeo} = \log_2\left[\frac{\text{Cn}}{1.5\text{Bn}}\right]$$

where Cn is the measured concentration of the element and Bn is the geochemical background value of the element or average shale. The constant 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic variations in the sediments (Lu et al. 2009). In this study, the background concentration of trace metals in the earth's crust was used as a reference value (Dasaram et al. 2011).

Contamination factor (CF)

To assess the extent of contamination of heavy metals and also to provide a measure of the degree of overall contamination along a particular route, contamination factor has been applied (Hakanson1980; Moore et al. 2009) and contamination factor (CF) is expressed as:

$$\mathrm{CF} = \frac{\mathrm{Cmetal\,(Sample)}}{\mathrm{Cmetal\,(Background)}}$$

where CF is the contamination factor, Cmetal is the concentration of pollutant in sediment, and Cmetal (Background) is the background value for the metal. The CF reflects the metal enrichment in the sediment. The geochemical background values in continental crust averages of the trace metals under consideration reported by Taylor and McLennan (1985) were used as background values for the metal concentration.

Statistical and geostatistical analyses

The descriptive statistics for the parameters analysed were calculated, and the correlations between heavy metals were assessed by using Pearson correlation analysis using SPSS 16.0 version for Windows. The positions of soil sample locations were recorded as a coordinate system using a GPS receiver. GPS surveying instrument Trimble Juno3 series equipped with a GPS receiver, antenna and rugged hand-held controller was used for positioning of sampling sites. The Trimble system brings precise subcentimetre control of elevation (Sankaran et al. 2012). Ordinary kriging interpolations of the contaminant concentrations in soil were computed with the Arc GIS 9.2 (Zawadzki and Fabijanczyk 2013).

Results and discussion

In order to assess petroleum hydrocarbon and heavy metal contamination in the soil adjacent to highways, samples were collected from ten automobile workshops from Gorimedu area of Pondicherry beside NH66 and six samples were collected from agricultural fields along NH45A and SH49 (Fig. 1). Experimental soil possessed the following physicochemical characteristics: sandy clay in texture (Water Year 2007), electrical conductivity (dS/m) was 0.21, soil pH ranged from 5.65 to 7.5, soil moisture ranged from 7.08 to 50.43, concentration of sodium ion (ppm) was 148–283, and potassium ion ranged from 168.00 to 271.04 ppm. The eastern part of Tamil Nadu and Pondicherry and Karaikkal were marked by a coastal plain with landforms such as vast tidal flats, continuous beach ridges, estuaries and lagoons and a narrow but fairly continuous beach. The coastline of Tamil Nadu and Pondicherry comprises a number of cusps, spits and wave-cut platforms and several palaeo-shorelines (GSI 2006).

Average concentrations of heavy metals

Soil samples from all the sites under investigation revealed varying concentrations of all the five analysed heavy metals (Pb, Zn, Cu, Mn, Cr and Cd). Metal concentration in the range of 143.07–319.28 mg kg^{-1} Cu; 68.72–396.41 mg kg^{-1} Pb; 162.42–284.91 mg kg^{-1} Zn; 212.72–401.33 mg kg^{-1} Mn; 0.12–15.41 mg kg^{-1} Cr; and 0.73–1.06 mg kg^{-1} Cd in dry soil was observed in the sampling site close to automobile workshops. However, in agricultural fields, the metal concentration varied between 33.68 and 66.62 mg kg^{-1} Cu; 27.22–73.66 mg kg^{-1} Pb; 26.24–75.59 mg kg^{-1} Zn; 137.88–242.07 mg kg^{-1} Mn; 0–0.21 mg kg^{-1} Cr; and 1.04–1.58 mg kg^{-1} Cd (Table 1). The concentration of heavy metals was found to be much lesser in soil from industrial areas and agricultural sites compared to those from automobile workshops. The selected relatively uncontaminated control site (Pondicherry University garden soil) showed a concentration of 29.84 mg kg^{-1} of Pb, 37.24 mg kg^{-1} Zn, 23.93 mg kg^{-1} Cu, 133.26 mg kg^{-1} of Mn and 1.46 mg kg^{-1} of Cd while Cr was not detectable in dry soil (Table 2). The results from agricultural soil corroborate with the works of Sankaran et al. (2012) reported from surface soil of SIPCOT. Roadside soils showed a high degree of contamination that can be attributed to motor vehicles. Several researchers have stated that the concentrations of the metals Pb, Cu, Zn, Cd and Ni decrease rapidly within 10–50 m from the roadside (Mmolawa et al. 2011; Mahbub et al. 2009).

The concentrations of all the studied metals in automobile workshops were higher compared to agricultural soil samples and control. The concentration of metals reported in our studies could be compared with surface soil heavy metal levels reported from other cities (Table 3). In urban soils and road dust, heavy metals were mainly released from traffic and industries while in agricultural soils, the sources of heavy metals were influenced by parent materials, mining, application of fertilizers and pesticides (Wei and Yang 2010).

The average concentration of Pb in the surface soil of automobile workshops beside NH66 was observed to be higher than other soil samples ranging between 68.72 and 396.41 mg kg^{-1}. Highest average concentration of 396.41 mg kg^{-1} lead was recorded in S3 which exceeds Canadian residential soil quality guidelines (CEQG 2002), but these levels were within the limits set by European Union (2002). Major sources of Pb in the atmosphere are combustion of petrol containing Pb additives, smelting, vehicular exhaust emissions related to urban transportation

Table 1 A description of sampling sites and average concentrations of heavy metals

Longitude (decimal)	Latitude (decimal)	Sample ID	National Highway	Cu	Pb	Zn	Mn	Cr	Cd	TPH
79.79423	11.96119	S1	NH66	233.15	167.14	228.97	401.33	13.82	1.06	90.72
79.79412	11.96154	S2	NH66	165.12	136.52	262.08	286.86	11.34	0.73	121.80
79.79399	11.96138	S3	NH66	143.07	396.41	214.17	242.04	13.44	0.86	118.25
79.79374	11.96161	S4	NH66	176.25	192.79	226.88	272.18	4.84	0.78	113.51
79.79379	11.96212	S5	NH66	245.22	134.72	208.04	266.41	15.41	0.77	116.25
79.79481	11.96165	S6	NH66	319.28	147.13	284.91	263.46	0.7	0.80	116.46
79.7951	11.96170	S7	NH66	216.12	68.72	166.22	212.72	2.26	0.87	108.37
79.79569	11.96366	S8	NH66	185.34	133.28	273.18	245.64	0.12	0.84	93.11
79.79567	11.96046	S9	NH66	193.96	112.99	197.70	254.85	2.11	0.95	108.71
79.79594	11.96086	S10	NH66	144.86	76.57	162.42	278.56	1.39	0.79	116.59
79.75845	11.69764	S11	NH45A	46.22	39.37	75.59	178.32	0.21	1.06	60.44
79.75751	11.69741	S12	NH45A	45.26	27.22	26.64	197.22	ND	1.04	70.22
79.76837	11.79151	S13	NH45A	37.98	62.72	33.99	274.07	ND	1.11	83.40
79.89489	12.09397	S14	SH49	63.62	59.57	26.24	158.45	ND	1.58	77.44
79.89489	12.09395	S15	SH49	43.59	61.3	36.15	189.14	ND	1.08	44.94
79.86983	12.04593	S16	SH49	33.68	73.66	26.28	137.88	ND	1.16	51.61

The values are given in (mg kg^{-1})

ND not detectable

Table 2 Descriptive statistics of the sampling sites in the study area

Area	Statistics	Cu	Pb	Zn	Mn	Cd	Cr	TPH
Automobile workshops (NH66)	Minimum	143.07	68.72	162.42	212.72	0.73	0.12	90.72
	Maximum	319.28	396.41	284.91	401.33	1.06	15.41	121.79
	Mean	202.24	156.63	222.46	272.40	0.85	6.54	110.38
	SD	53.64	92.18	41.85	49.99	0.10	6.19	10.56
	Variance	2,877.12	8,497.18	1,751.18	2,499.18	0.01	38.35	111.51
Agricultural fields (NH45A, SH49)	Minimum	33.68	27.22	26.24	137.88	1.04	ND	44.94
	Maximum	63.62	73.66	75.59	274.07	1.58	0.21	83.4
	Mean	45.06	53.97	37.48	189.18	1.17	0.03	64.68
	SD	10.27	17.19	19.16	46.83	0.21	0.09	14.98
	Variance	105.55	295.61	367.08	2,192.92	0.04	0.01	224.45

(Khan et al. 2011; Khillare et al. 2004). Approximately 75 % of Pb contained in leaded gasoline gets emitted directly into the atmosphere. Only 25 % of Pb released by vehicles being in coarse fraction gets deposited close on roads, and the remaining fine fraction remains airborne contaminating areas more remote from the point of its emission (Fergusson and Kim 1991). Lead deposited in the road dust and vehicular traffic in urban soils having less mobility stays on the surface soil. So, there is a lesser possibility to enter into roots of higher plants and may not be transported to the rest of the plant parts. Only shallow-rooted plants are much vulnerable to picking up lead contamination (Bentum et al. 2011).

The average concentration of Zn was 222.46 mg kg^{-1}, and it varied between 162.42 to 284.91 mg kg^{-1} in workshop area. These values were high compared to those found in agricultural fields (26.24–75.59 mg kg^{-1}). Zn used in brake linings because of their heat-conducting properties could be released from combustion of engine oil and tyres of motor vehicle during mechanical abrasion (Christiana and Samuel 2013). Zn could interrupt the biochemical activities in soils by retarding the growth of microorganisms, and earthworms (Wuana and Okieimen 2011) are considered phytotoxic in elevated concentrations, directly affecting crop yields and soil fertility (Bentum et al. 2011). Liu et al. (2013) reported Zn concentration in

Table 3 Comparison of average concentrations of heavy metals in roadside surface soil from different highways connected to cities

Locations	Cu	Pb	Zn	Mn	Cd	Cr	References
Pondicherry NH66	202.24	156.63	222.46	272.40	6.54	0.85	Present study
ECR-NH45A and SH 49	45.06	53.97	37.48	189.18	1.17	0.03	Present study
Canadian standards	63	140	200	–	10	64	CEQG (2002)
European Union standards	140	300	300	–	3.0	150	EU (2002)
Indian standard	135–270	250–500	300–600	–	3–6	–	Awashthi (2000)
Assam Trunk Road-NH37 Guwahati, Assam	103.9–238.8	110–285.5	166.8–697.2	321.8–512.2	3.1–15.9	98.9–128.6	Mahanta and Bhattacharyya (2011)
NH24 and NH25	–	60.65–16.66	–	–	–	–	Singh et al. (1997)
Cossipore, Kolkata	–	118	529	3	5	6	Karar et al. (2006)
Karnal Bypass, Delhi	128.98–307.1	187.06–292.94	241.73–420.78		17.92	171.80–764.34	Anju and Banerjee (2005)
Highways 400, 401 and 404 Greater Toronto area, Canada	162	182.8	200.3	1,202.2	0.51	197.9	Nazzal et al. (2013)
214# National Highway, 308# Provincial Highway, 109# National Highway, China	23.13	29.60	100.97	–	0.30	36.20	Yan et al. (2013)
Arniko Highway, Nepal	–	245.36	–	–	2.84	–	Raj and Ram (2013)
Murree Highway, Islamabad, Pakistan	156.9	145.8	890	–	8.4	93	Abbasi et al. (2013)

The values are given in (mg kg^{-1})

soil collected from Chaoyangas fluctuating from 22.787 to 669.597 mg/kg showing slight contamination as supported by geoaccumulation index.

The average concentration of Cu was 198.81 ± 167.66 mg kg^{-1} ranging between a minimum of 136.64 mg kg^{-1} to a maximum value of 319.28 mg kg^{-1} in workshops while in agricultural fields, the concentrations varied between 33.68 and 66.62 mg kg^{-1}. Break dust recognized as a carrier of Cuis used in brakes to control heat transport (Christiana and Samuel 2013). Agricultural soils showing elevated levels of Cu might have reached the contaminant levels by application of pesticides. Cu translocated to crops becomes hazardous for human consumption (Bentum et al. 2011). Manganese showed an average mean of 270.59 ± 164.09 mg kg^{-1} ranging from 401.33 to 212.72 mg kg^{-1}. The concentration of manganese did not vary much in both the sampling areas.

Street dust generally contained low concentration of Cd when compared to all other metals in consistence with the findings of Faiz et al. (2009). The average concentration of Cd was 0.845 ± 1.42 mg kg^{-1}, and it varied between 0.73 and 1.06 mg kg^{-1} Cd along NH66 while in agricultural fields, the recorded concentration was 1.04–1.58 mg kg^{-1}. Raj and Ram (2013) reported a cadmium mean value of 2.84 mg/kg across the road section of the Kathmandu–Bhaktapur road section of Arniko Highway. The possible sources of high Cd contamination include slow destruction of automobile tires, car abrasion, lubrication oils and galvanized parts of vehicles. Cadmium concentrations range from 0.07 to 0.10 ppm in diesel oils and 0.20–0.026 ppm in lubricating oils. Cadmium accumulates in agricultural soils through the application of soil amendments like phosphatic fertilizers and sewage sludges (Ramachandran and D'Souza 1998). The average concentration of Cr in workshops found was 6.54 ± 11.43 mg kg^{-1} ranging from a minimum of 0.12 mg kg^{-1} to a maximum of 15.41 mg kg^{-1}, whereas 0–0.21 mg kg^{-1} Cr was found in agricultural areas sampled. It finds entry into the environment through natural sources such as bio cycling, weathering of rocks and anthropogenic input of Cr comes from solid wastes, where approximately 30 % of Cr originates from disposal of plastic bags (Vandana et al. 2011).

Total petroleum hydrocarbons

Total petroleum hydrocarbon (TPH) concentration ranged between 90.72 and 121.79 mg kg^{-1} in automobile workshop contaminated soil, 44.94–83.4 mg kg^{-1} in agricultural sites while 4.55 mg kg^{-1} concentration of TPH was present in control at a depth of 0–15 cm from the surface.

All sites showed higher concentration of total petroleum hydrocarbons than the control, which is 3 km away from SH49 and less exposed to atmospheric and roadside dust. The highest mean TPH level was observed at site 2 (121.79 mg kg^{-1}) (Table 1).

Our findings corroborate with TPH concentration for the top soils measured at 0–15 cm depth ranging from 55 ± 13 to 302 ± 14 mg kg^{-1} from the Niger Delta region of Nigeria 3 months after an extensive oil spillage (Okop and Ekpo 2012). In automobile waste dumps total petroleum hydrocarbon levels in all sites ranged from 486 to 4,438.7 mg kg^{-1} at 0–15 cm depth (Chukwujindu et al. 2008). Adeniyi and Afolabi (2002) reported TPH concentrations at petrol stations having minimum of 399.83 ± 106.19 and maximum of 450.83 ± 90.58 µg/g, respectively, mechanic workshops, 362.60 ± 185.84 and 428.55 ± 119.00 µg/g, respectively, while the National Electric Power Authority (NEPA) station reported 356.20 ± 210.30 µg/g as compared to the control mean of 26.63 ± 4.58 µg/g. In soil samples collected from a garage, near Crawford University, Igbesa, Ogun state, the sites contained mean TPH values of 19.43 ± 1.27, 16.11 ± 1.85 and 11.43 ± 4.33 mg/g (Adeleke et al. 2010). Much higher levels of TPHs in the order of 1,179.3 to 6,354.9 mg kg^{-1}, with the average of 2,676.6 mg kg^{-1}, were reported from agricultural soils adjacent to petrochemical complex in Guangzhou, the capital city of Guangdong Province in southern China (Li et al. 2012a, b).

Spatial analysis using GIS

GIS software is increasingly used in environmental studies because of its ability to interpret spatial distribution of trace metals (Monitha et al. 2012). The spatial distribution and variability of contaminants in soils were analysed, and metal concentrations were interpolated with the ordinary kriging method (Li et al. 2012a, b) as it is useful tool to assess the possible sources of enrichment and to identify hot spot area with high metal concentration (Viguri et al. 2007). The geochemical maps of Pb, Zn, Cu, Mn, Cd and Cr are presented in Fig. 2 in which hot spots of high metal concentrations were identified in the geochemical maps. Similar spatial distribution patterns of Pb, Zn, Cu, Mn, Cd and Cr were found in the geochemical maps. This provides confirmation of the results in the statistical analysis, where the distribution of contaminants was found to be higher in the highways adjacent to workshops. Strong associations were found among these metals with total petroleum hydrocarbons.

Geoaccumulation index (Igeo)

There is no documented information available on typical background values for automobile workshop sediments of Puducherry for heavy metal concentrations; therefore, these data were compared with world average shale values as background values (Gibbs 1993).

The calculated Igeo values, based on the world average shale values as background values (Rubio et al. 2000) for the sampling sites, were illustrated in Table 4. Table 5 gives an account of pollution classes based on studies of Muller (1969). The pollution levels of the analysed metals at the study site expressed in terms of geoaccumulation indices indicated that the soil at the sites was not polluted with regard to Cr and Mn; Cu (S11, S12, S13 and S14). Agricultural areas were non-polluted to moderately polluted (class 1) with Zn, Cu (S15, S16) and Pb (S11 and S14). Cd, Zn and Cu (except S6) in automobile workshop sites; Pb (S7, S9, S10, S13, S15 and S16) belonged to class 2 pollutants. S1, S2, S4, S5, S6, S8 of automobile workshop sites were moderately polluted to polluted (class 3) with Pb. Site S3 fell under the category of contaminated to heavily contaminated (class 4) with Pb. Wei and Yang (2010) reported Igeo values in agricultural soils of the 12 cities from China which were lower than zero. These findings indicate that the agricultural soils are uncontaminated or slightly contaminated by the metals.

Contamination factor

Contamination factors of various metals in the soils from sampled sites were presented in Table 6. Based on the contamination factor categories, all sampling sites showed considerable to very high contamination of Pb. Sites 1, 5 and 6 showed very high contamination with Pb where sampling site 6 showed a maximum of ($CF = 7.095$) contamination. Considerable contamination factor was recorded in case of Cu (Table 7). Moderate contamination of Zn and a low contamination of Mn, Cd and Cr were also recorded. The general trend of mean contamination factor was $Pb > Zn > Cu > Mn > Cr > Cd$. Topalian et al. (1999) reported an association among Pb, Zn and Cu and with its significance being independent of sampling station. Lead and other heavy metals such as zinc, cadmium, copper, manganese and nickel were the additives in some of the lubricants and gasoline and were non-degradable in the soil (Ipeaiyeda and Dawodu 2008). Various indices showed that areas located nearby automobile workshops were highly contaminated with Pb in the present study. The agricultural areas, though invariably enriched with these toxic metals, showed comparatively less contamination compared to soil from automobile workshops possibly due to uptake by plants and also as they distantly located from areas exposed to direct disposal of contaminants. The primary risk associated with uptake of heavy metals by crops is their ability to enter into different compartments of food chain. A high level of toxic elements in the tissues of crop

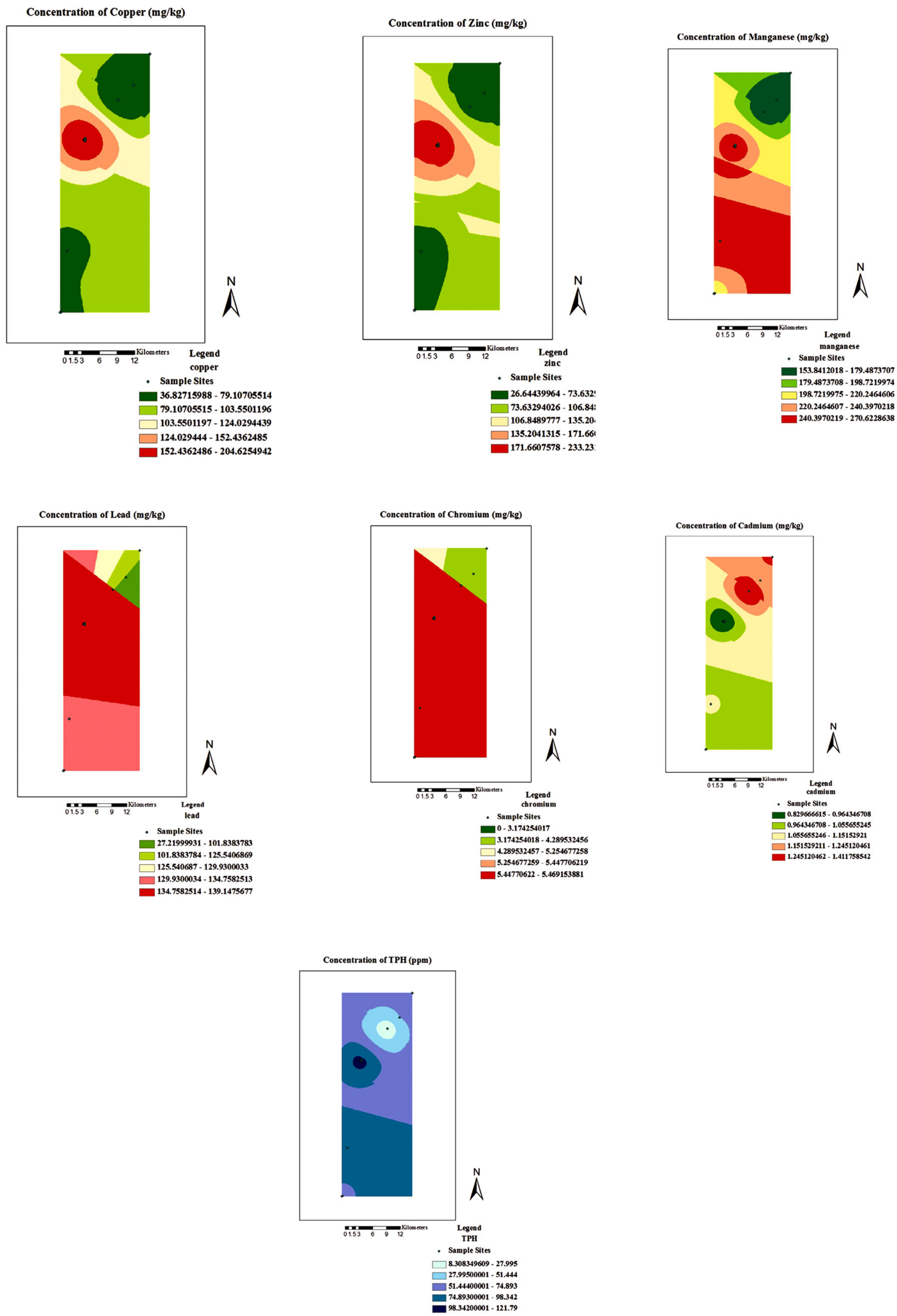

Fig. 2 Spatial variability of available concentration of heavy metals and TPH in soil

Table 4 Geoaccumulation index of the samples for heavy metals in the soil of the study sites

Sample ID	Cu	Pb	Zn	Mn	Cd	Cr
S1	1.789	2.478	1.096	−1.748	1.685	−9.910
S2	1.292	2.18	1.123	−2.233	1.287	−10.374
S3	1.085	3.724	1.082	−2.478	1.462	−11.89
S4	1.386	2.684	1.094	−2.309	1.382	−10.55
S5	1.862	2.167	1.076	−2.340	1.361	−10.278
S6	2.243	2.294	1.140	−2.356	1.412	−14.234
S7	1.680	1.196	1.031	−2.665	1.528	−11.931
S8	1.458	2.152	1.131	−2.457	1.481	−10.175
S9	1.524	1.914	1.066	−2.404	1.546	−11.144
S10	1.103	1.352	1.026	−2.276	1.392	−11.137
S11	−0.382	0.392	0.872	−2.919	1.235	−11.350
S12	−0.457	−0.140	0.662	−2.774	1.204	ND
S13	−0.442	1.064	0.711	−2.299	1.291	ND
S14	−0.084	0.990	0.659	−3.090	1.812	ND
S15	0.329	1.031	0.723	−2.834	1.265	ND
S16	0.660	1.296	0.659	−3.290	1.365	ND

Table 5 Threshold values of geoaccumulation index

Igeo value	Class	Pollution class
Igeo < 0	0	Practically uncontaminated
0 < Igeo < 1	1	Uncontaminated to moderately uncontaminated
1 < Igeo < 2	2	Moderately contaminated
2 < Igeo < 3	3	Moderately to heavily contaminated
3 < Igeo < 4	4	Heavily contaminated
4 < Igeo < 5	5	Heavily to extremely contaminated
5 < Igeo	6	Extremely contaminated

Source: Muller (1969)

plants causes a reduction in biomass and yields (Bentum et al. 2011).

Correlation between heavy metals and total petroleum hydrocarbons

Pearson correlation analysis was performed to assess possible sources of contamination and the inter-relationships existing among variables depicted in the form of a correlation matrix. Heavy metals in soil usually have complicated relationships among them (Sun et al. 2010). Total petroleum hydrocarbon concentration shows strong positive correlation with Zn ($r = 0.811$), Cu ($r = 0.761$); Pb ($r = 0.642$), Mn ($r = 0.571$), Cr ($r = 0.530$) and strong negative correlation with Cd ($r = -0.765$) at 0.05 significance level (Table 8). Correlation between TPH and heavy metals Zn, Cu and Pb suggests that materials released from common source might have contributed to contamination

Table 6 Contamination factors of heavy metals for the study sites

Sample ID	Cu	Pb	Zn	Mn	Cd	Cr
S1	4.567	6.047	5.866	1.973	1.036	3.115
S2	3.234	4.939	6.715	1.410	0.786	2.259
S3	2.803803	14.342	5.487	1.190	0.887	0.790
S4	3.453	6.975	5.813	1.338	0.839	2.00
S5	4.803	4.874	5.330	1.310	0.827	2.411
S6	6.254	5.323	7.300	1.295	0.857	0.156
S7	4.233	2.486	4.259	1.046	0.929	0.768
S8	3.631	4.822	6.999	1.208	0.899	2.593
S9	3.799	4.088	5.065	1.253	0.940	1.325
S10	2.838	2.77	4.162	1.369	0.845	1.331
S11	1.014	1.424	1.937	0.877	0.758	1.148
S12	0.963	0.985	0.683	0.970	0.742	0
S13	0.972	2.270	0.871	1.347	0.788	0
S14	1.246	2.155	0.672	0.779	1.131	0
S15	1.659	2.218	0.926	0.930	0.774	0
S16	2.088	2.665	0.673	0.678	0.829	0

Table 7 Estimation of contamination factor

CF range	Contamination level
CF < 1	Low contamination
1 ≤ CF < 3	Moderate contamination
3 ≤ CF ≤ 6	Considerable contamination
CF > 6	Very high contamination

Source: Moore et al. (2009)

(Thavamani et al. 2012). Numerous factors like original contents of heavy metals in rocks and parent materials, various processes of soil formation and anthropogenic contamination contributing factors (Li et al. 2008) control their relative abundance. The high negative correlation value found between metals Cd and all other metals indicated that the resultant contamination might be due to different sources, as well as their different sinks in the soil profile (Matini et al. 2011). It could be assumed that the heavy metals analysed except Cd were derived almost exclusively from automobiles (Addo et al. 2012) and road transport activities associated with highways, corrosion from the possible sources such as junk metal items, scrapyards and auto-repair shops, street dust and storm water as a result of automobile traffic (Nava-Martinez et al. 2012).

Conclusion

The concentration of heavy metals beside NH66 close to automobile workshops ranged between 143.07 and 319.28 mg kg^{-1} Cu; 68.72–396.41 mg kg^{-1} Pb; 162.42–284.91 mg kg^{-1} Zn; 212.72–401.33 mg kg^{-1} Mn;

Table 8 Pearson correlation among selected heavy metals and total petroleum hydrocarbons

		Pb	Zn	Cu	Mn	Cd	Cr	TPH
Pb	Pearson correlation	1	0.591*	0.388	0.379	−0.401	0.659**	0.552*
	Sig. (2-tailed)	–	0.016	0.138	0.148	0.124	0.006	0.027
	N	16	16	16	16	16	16	16
Zn	Pearson correlation	0.591*	1.000	0.846**	0.656*	−0.751**	0.529*	0.811**
	Sig. (2-tailed)	0.016	–	0.000	0.006	0.001	0.035	0.000
	N	16	16	16	16	16	16	16
Cu	Pearson correlation	0.388	0.846**	1.000	0.554*	−0.605*	0.436	0.681*
	Sig. (2-tailed)	0.138	0.000	–	0.026	0.013	0.091	0.004
	N	16	16	16	16	16	16	16
Mn	Pearson correlation	0.379	0.656**	0.554*	1.000	−0.473	0.603*	0.583*
	Sig. (2-tailed)	0.148	0.006	0.026	–	0.064	0.013	0.018
	N	16	16	16	16	16	16	16
Cr	Pearson correlation	0.659**	0.529*	0.436	0.603*	−0.394	1.000	0.530*
	Sig. (2-tailed)	0.006	0.035	0.091	0.013	0.131	–	0.035
	N	16	16	16	16	16	16	16
Cd	Pearson correlation	−0.401	−0.751**	−0.605*	−0.473	1.000	−0.394	−0.687**
	Sig. (2-tailed)	0.124	0.001	0.013	0.064	–	0.131	0.003
	N	16	16	16	16	16	16	16
TPH	Pearson correlation	0.552*	0.811**	0.681**	0.583*	−0.687**	0.530*	1.000
	Sig. (2-tailed)	0.027	0.000	0.004	0.018	0.003	0.035	–
	N	16	16	16	16	16	16	16

* Correlation is significant at the 0.05 level (2-tailed)

** Correlation is significant at the 0.01 level (2-tailed)

0.12–15.41 mg kg^{-1} Cr; and 0.73–1.06 mg kg^{-1} Cd in dry soil. However, in agricultural fields, the concentrations varied between 33.68 and 66.62 mg kg^{-1} Cu; 27.22–73.66 mg kg^{-1} Pb; 26.24–75.59 mg kg^{-1} Zn; 137.88–242.07 mg kg^{-1} Mn; 0–0.21 mg kg^{-1} Cr; and 1.04–1.58 mg kg^{-1} Cd. The results showed that highways adjacent to automobile workstations were exposed to elevated levels of heavy metal and T PH contamination. The Igeo values indicated that the soil was unpolluted with Cr and Mn, unpolluted to moderately polluted with Zn and Cd (class 1); moderately polluted with Cu (class 2) and moderately polluted to polluted in the case of Pb (class 3); unpolluted to moderately polluted with Mn and Zn (class 1); moderately polluted with Cd and Cu (class 2) and moderately polluted to polluted in the case of Pb (class 3). The sites showed very high contamination of Pb, considerable contamination of Cu, moderate contamination of Zn and a low contamination of Mn, Cd and Cr in terms of contamination factor. Pearson correlation matrix also directs strong positive correlation between TPH and heavy metals (Cu, Zn and Pb), suggesting a common source for the positively correlated heavy metals. Using GIS spatial analysis methods, the spatial variability in soil contamination was manifested. The findings of the present study would be helpful for understanding soil contamination in the surface soil near national highways influenced by road transportation. If these metals leak into adjacent agricultural fields, they slowly find entry into food chain leading to serious health hazards. Therefore, there is an urgent need for policy regulations to minimize indiscriminate disposal of oil contaminated residues, vehicular emissions, road transport and traffic emissions and the wear and tear of mechanical parts in vehicles beside urban highways.

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