Journal of Environmental Management 220 (2018) 198–206

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Health risk assessment of heavy metals in atmospheric deposition in a congested city environment in a developing country: Kandy City, Sri Lanka

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A R T I C L E I N F O

Article history:
Received 30 May 2017
Received in revised form
5 April 2018
Accepted 7 April 2018

Keywords:
Heavy metals
dry deposition
Atmospheric pollution
Human health risk
Sri Lanka

A B S T R A C T

This research study which was undertaken in a congested city environment in a developing country provides a robust approach for the assessment and management of human health risk associated with atmospheric heavy metals. The case study area was Kandy City, which is the second largest city in Sri Lanka and bears the characteristics of a typical city in the developing world such as the urban footprint, high population density and traffic congestion. Atmospheric deposition samples were collected on a weekly basis and analyzed for nine heavy metals common to urban environments, namely, Al, Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb. Health risk was assessed using hazard quotient (HQ) and hazard index (HI), while the cancer risk was evaluated based on life time daily cancer risk. Al and Fe were found to be in relatively high concentrations due to the influence of both, natural and anthropogenic sources. High Zn loads were attributed to vehicular emissions and the wide use of Zn coated building materials. Contamination factor and geo-accumulation index showed that currently, Al and Fe are at uncontaminated levels and other metals are in the range of uncontaminated to contaminated levels, but with the potential to exacerbate in the long-term. The health risk assessment showed that the influence of the three exposure pathways were in the order of ingestion > dermal contact > inhalation. The HQ and HI values for children for the nine heavy metals were higher than that for adults, indicating that children may be subjected to potentially higher health risk than adults. The study methodology and outcomes provide fundamental knowledge to regulatory authorities to determine appropriate mitigation measures in relation to HM pollution in city environments in the developing world, where to-date only very limited research has been undertaken.

1. Introduction

Atmospheric pollution is a serious problem in most cities around the world as it has a direct impact on human and ecosystem health (Gunawardena et al., 2013b). Among the various atmospheric pollutants, particulates are of significant concern because of their association with other chemical and microbial pollutants (Amodio et al., 2014; Anatolaki and Tsitouridou, 2007; Huang et al., 2009; Weerasundara et al., 2017). The atmospheric particulates can be either natural or anthropogenic in origin (Bermudez et al., 2012; Soriano et al., 2012). Natural atmospheric particulates primarily originate from roadside soil due to wind and vehicle related turbulence (Shi et al., 2008; Weerasundara et al., 2017), while anthropogenic particulates are generated by industrial activities, agricultural activities, domestic emissions, as well as automobile activities such as vehicle exhaust, tyre wear, brake wear and road pavement wear (Soriano et al., 2012; Wei and Yang, 2010; Liu et al.,

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https://doi.org/10.1016/j.jenvman.2018.04.036
0301-4797/

2018). These particulates are removed from the atmosphere through wet (precipitation scavenging) and dry deposition processes by turbulent vertical transport (Connan et al., 2013).

Heavy Metals (HMs) attached to atmospheric particulates are considered as a potential threat to human health as these can be inhaled, ingested and/or contacted via dermal pathway and accumulate in the fatty tissues and affect the central nervous system, or may act as cofactors in diseases (Huang et al., 2014; Lu et al., 2014; Ma et al., 2017). Potential sources of metals in the atmosphere are primarily vehicular and industrial activities (Connan et al., 2013; Weerasundara et al., 2017). Being non-degradable, significant levels of HMs can threaten the ecosystem through stormwater pollution (Gunawardena et al., 2013a; Ma et al., 2016). Past literature on atmospheric deposition has primarily focused on the concentration, distribution and source identification (Connan et al., 2013; Egodawatta et al., 2013; Soriano et al., 2012; Wei et al., 2009). Only limited studies have focused on the assessment of their potential health risks, despite health risk assessment being an essential foundation of risk management (Wei et al., 2015).

In Kandy, Sri Lanka, atmospheric pollution has become a serious issue during the past decade (Ileperuma, 2010). As Kandy is the second largest city in Sri Lanka, it can exert significant influence on the Country's economy. More than ten schools are located in the heart of the highly congested Kandy City. Children have increased vulnerability to atmospheric particulates due to the nature of their daily activities (USEPA, 2008). Kandy also has religious and historical importance and is listed as a UNESCO World Heritage City. Therefore, Kandy attracts a large number of local visitors, and foreign tourists, who could be affected by the polluted atmosphere. Kandy bears the characteristics of a typical city in the developing world such as high population density and traffic congestion. The major pollution source in the City is attributed vehicular emissions as industrial activities are very limited within the metropolitan area. A study undertaken in 2011 (Wickramasinghe et al., 2011), confirmed a daily traffic volume of 106,000 vehicles within the Kandy City limits. It is also important to note that this traffic volume is confined to a relatively limited land area of 4 km^2 (Wickramasinghe et al., 2011). Additionally, a transient population of about 100,000 visit the City on a daily basis, resulting in a total population of more than 250,000 (Wickramasinghe et al., 2011).

Studies have been undertaken to assess atmospheric pollution through atmospheric deposition (Duan and Tan, 2013; El-Araby et al., 2011; Huang et al., 2014). Studies on atmospheric deposition of HMs and related risk assessments are limited in the context of a typical city in a developing country, particularly in the Sri Lankan context. This study was conducted to assess the human health impacts posed by HMs associated with atmospheric deposition in Kandy City and its environs. The study results provide important information to regulatory authorities to determine appropriate mitigation measures in relation to HM pollution of Kandy City atmosphere.

2. Materials and methods

2.1. Study area and sampling sites

Kandy is located at approximately 500 m above mean sea level. The average day time ambient temperature is in the range of 28–32 °C, monthly rainfall is in the range of 52–398 mm and the daytime relative humidity is in the range of 63–83%. The City is located in a valley surrounded by mountains, facilitating thermal inversions within the city atmosphere. Kandy is 26 km^2 in extent with a permanent population of more than 170,000 and a daily transient population of around 100,000 (Wickramasinghe et al., 2011). The City has four main entry points and the high daily traffic flow and the limited land area as noted above, results in high traffic congestion and exceptionally high vehicular emissions (Wickramasinghe et al., 2011).

The sampling sites were spread around Kandy and its environs. Nine sampling sites were selected considering different traffic characteristics. These sites were designated as Children's Park Station (C), Dodanwela site (D), Fire Brigade Station (F), National Institute of Fundamental Studies (I), Lewalla (L), Police Station (P), Railway Station (R), Trinity college site (TC), and Tea Research Institute (TRI) (Fig. S1). Sites C, F, P, R, and TC are in high traffic volume areas, while sites D and I are in low traffic volume areas. Sites L and TRI were selected as control sites as these two sites are located away from arterial roads and have relatively low vehicular and other anthropogenic activities.

2.1.1. Sample collection

Atmospheric deposition samples were collected weekly as dry deposition. The samplers were constructed using high density polyethylene bottles with polyethylene funnels, connected to a star picket bar, and fixed at a height of 1.5 m above ground to minimize contamination from re-suspended particles. The sample collection system was previously described by Weerasundara et al. (2017). Prior to installation, the sampling bottles and funnels were washed with deionized water followed by an acid wash with 1:1 HNO_3 solution as part of the quality assurance measures. At the end of each sample collection, sample bottles and funnels were replaced. After collection, the funnels were enclosed in clean plastic bags and sealed to avoid contamination. Sample bottles were also sealed and transported to the laboratory immediately following standard quality control procedures. The dry deposition samples were collected on a weekly basis and the sample collection was undertaken for ten weeks (ten dry deposition samples). The samples affected by rainfall were discarded.

2.2. Laboratory analysis of heavy metals in atmospheric deposition

After the samples were brought to the laboratory, the funnels and bottles were washed with autoclaved deionized water in order to transfer samples to polyethylene bottles. The samples were stored at 4 °C temperature under >2 pH until laboratory analysis was carried out. The preserved 50 mL samples were digested with 1:1 HCl and 1:1 HNO_3 acid solutions, in a water bath at a temperature of <80 °C until the volume reached 20 mL (USEPA, 1994). The HM concentrations were determined according to Method 200.8 (USEPA, 1994) using an Agilent 8800 Triple Quadruple Inductively Coupled Plasma Mass Spectrometer (ICP-MS). The quality assurance and quality control (QA/QC) samples were prepared and tested as specified in US EPA Method 200.8 (USEPA, 1994).

2.3. Contamination assessment

Metal concentration in a given environment is influenced by a range of factors such as the nature of the substrate, environmental conditions, availability of pollution sources, and distance from emission sources (Nobi et al., 2010). Contamination factor (CF) is a tool that can be used to determine the pollution status of a given environment over a period of time (Nobi et al., 2010; Varol, 2011). Using Eq. (1), CF was calculated to determine whether a particular site is polluted with HMs (Nobi et al., 2010; Varol, 2011).

$$CF = \frac{C_n}{B_n} \quad (1)$$

where, C_n is the concentration of contaminant in dust, and B_n is the background concentration for the particular contaminant.

A CF value > 1 for a particular metal indicates that the site is contaminated by the metal, while CF < 1 suggests that there is no metal enrichment by outside sources (Nobi et al., 2010). Similarly, it is important to assess the level of contamination by a particular metal. Accordingly, the geo-accumulation index (I_{geo}) is used to assess the contamination levels. Equation (2) was used to determine I_{geo} (Ma and Singhirunnusorn, 2012; Zheng et al., 2015).

$$I_{geo} = \log_2\left[\frac{C_n}{1.5B_n}\right] \tag{2}$$

C_n is the concentration of a particular metal in atmospheric deposition and B_n is the geochemical background concentration of the HM (crustal average). A constant of 1.5 is introduced to minimize the effect of possible variations in the background values (Ma and Singhirunnusorn, 2012; Zheng et al., 2015).

The I_{geo} is classified into the following 7 classes to categorize the HM pollution levels in atmospheric deposition (Ma and Singhirunnusorn, 2012; Zahra et al., 2014; Zheng et al., 2015).

a. $I_{geo} \leq 0$ - class 0, uncontaminated
b. $0 < I_{geo} \leq 1$ - class 1, uncontaminated to moderately contaminated
c. $1 < I_{geo} \leq 2$ - class 2, moderately contaminated
d. $2 < I_{geo} \leq 3$ - class 3, moderately contaminated to heavily contaminated
e. $3 < I_{geo} \leq 4$ - class 4, heavily contaminated
f. $4 < I_{geo} \leq 5$ - class 5, heavily contaminated to extremely contaminated
g. $5 < I_{geo}$ - class 6, extremely contaminated.

2.4. Health risk assessment

2.4.1. Exposure dose

Exposure to HMs in atmospheric deposition was assessed based on models developed by USEPA (2001). The average daily dose (ADD) of a particular HM via ingestion, inhalation and dermal contact pathways were calculated using Eqs. (3)–(5) (USEPA, 2001).

$$D_{ing} = C_n \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{3}$$

$$D_{inh} = C_n \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \tag{4}$$

$$D_{dermal} = C_n \times \frac{SL \times SA \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{5}$$

D_{ing} is the daily dose via ingestion of atmospheric deposition; D_{inh} is the daily dose via inhalation through mouth and nose; and D_{dermal} is the daily dose via dermal absorption of trace elements in deposition adhering to exposed skin. The definitions and values of the other parameters are given in Table S1.

Lifetime average daily dose of cancer elements (LADD) was calculated for the inhalation route using Eq. (6) (Ma and Singhirunnusorn, 2012).

$$LADD = \frac{C_n \times EF}{AT \times PEF} \times \left(\frac{InhR_{child} \times ED_{child}}{BW_{child}} + \frac{InhR_{adult} \times ED_{adult}}{BW_{adult}}\right) \tag{6}$$

2.4.2. Risk assessment

The Hazard Quotient (HQ) was calculated to assess the non-cancer risk posed by the nine metals. The ADD for the three exposure pathways was divided by the specific reference dose (RfD) (mg/kg/day) for a particular metal to obtain the HQ (Du et al., 2013; Ma and Singhirunnusorn, 2012; USEPA, 2001) (Eq. (7)). The RfD threshold determines the possibility of occurrence of health effects during a life time due to a particular pollutant (Du et al., 2013). If ADD < RfD, the HQ value will be lower than 1. HQ < 1 suggests that there is no possibility of occurrence of any adverse health effect due to the specific pollutant. If the HQ > 1, most probably the exposure pathway will cause adverse human health impacts (Du et al., 2013).

The hazard index (HI) is the sum of HQs (Eq. (8)). HI is used to assess the cumulative non-cancer risk from a single metal (Du et al., 2013; Lu et al., 2014; Ma and Singhirunnusorn, 2012). Similar to the interpretation of HQ, if HI < 1, it is considered that there is no significant risk from non-carcinogenic effects. If HI > 1, there is a possibility for non-carcinogenic effects to occur, with the probability tending to increase as HI increases (Du et al., 2013; Lu et al., 2014; USEPA, 2001).

$$HQ = \frac{ADD}{RfD} \tag{7}$$

$$HI = \sum_{i=1}^{3} HQ_i \tag{8}$$

To assess the cancer risk from carcinogenic metals, the calculated daily dose was multiplied by the corresponding slope factor (SF) (mg/kg/day) for a particular carcinogen (Du et al., 2013; Lu et al., 2014; USEPA, 2001).

3. Results and discussion

3.1. Heavy metal concentrations

The average HM concentrations in atmospheric deposition at the nine sampling sites are presented in Fig. 1. Al and Fe, which are present in significantly higher concentrations at all the sampling sites, were in the ranges of 507 - 43,328 and 770 - 43,922 mg/kg, respectively. The geogenic origin of these two metals would be the primary reason for the high concentrations (Bhuiyan et al., 2010; Ziyath et al., 2016).

The results of EF assessment also suggest that Al is of crustal origin. However, these metals could be re-suspended by disturbed soils through anthropogenic activities such as traffic, and eventually deposit on ground surfaces (Weerasundara et al., 2017). Other than Al and Fe, Zn was found in relatively higher concentration than the rest of the metals with the average being in the range of 137–3331 mg/kg. Zn is a major constituent in lubrication oil additives, tyre and brake wear, and galvanized automobile components (Duan and Tan, 2013).

Additionally, the wide use of Zn coated roofing materials and galvanized building components could also be attributed to the Zn loads in atmospheric deposition (Duan and Tan, 2013). Although, leaded gasoline has been phased out, the roadside soil could be enriched with Pb from past usage of leaded fuel and disturbance from traffic activities could result in re-suspension in the atmosphere (Egodawatta et al., 2013). Other HMs were reported in relatively low concentrations in weekly atmospheric deposition samples and decreased in the order, Mn > Cu > Cr > Ni > Cd. Spatial distribution of HMs in dust samples are presented in Fig. 2.

The spatial distribution maps were created using ArcMap 10.2.1 GIS software. Interpolated raster data with coordinates of 80° 37′ 55″ E, 7° 18′ 25″ N in the upper left corner, and 80° 39′ 04″ E, 7° 17′ 05″ N in the lower right corner were employed. Kriging interpolation was used for the raster interpolation from the point source

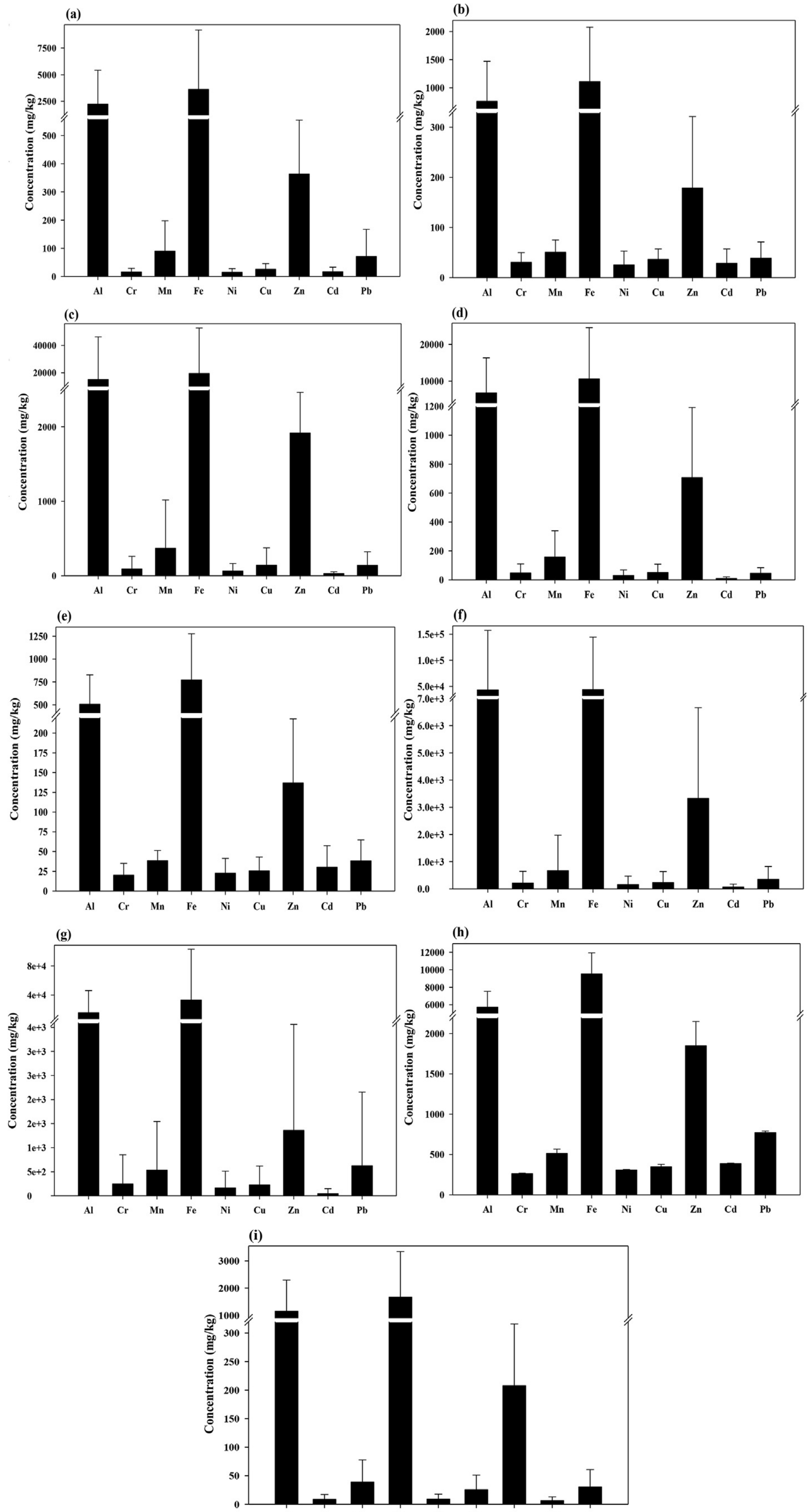


Fig. 1. Heavy metal concentrations at the nine sampling locations in Kandy and its environs. (a) Children's park (site C), (b) Dodanwela (site D), (c) Fire brigade station (site F), (d) National Institute of Fundamental Studies (site I), (e) Lewalla (site L), (f) Police station (site P), (g) Railway station (Site R), (h) Trinity college station (site TC), and (i) Tea Research Institute (site TRI).

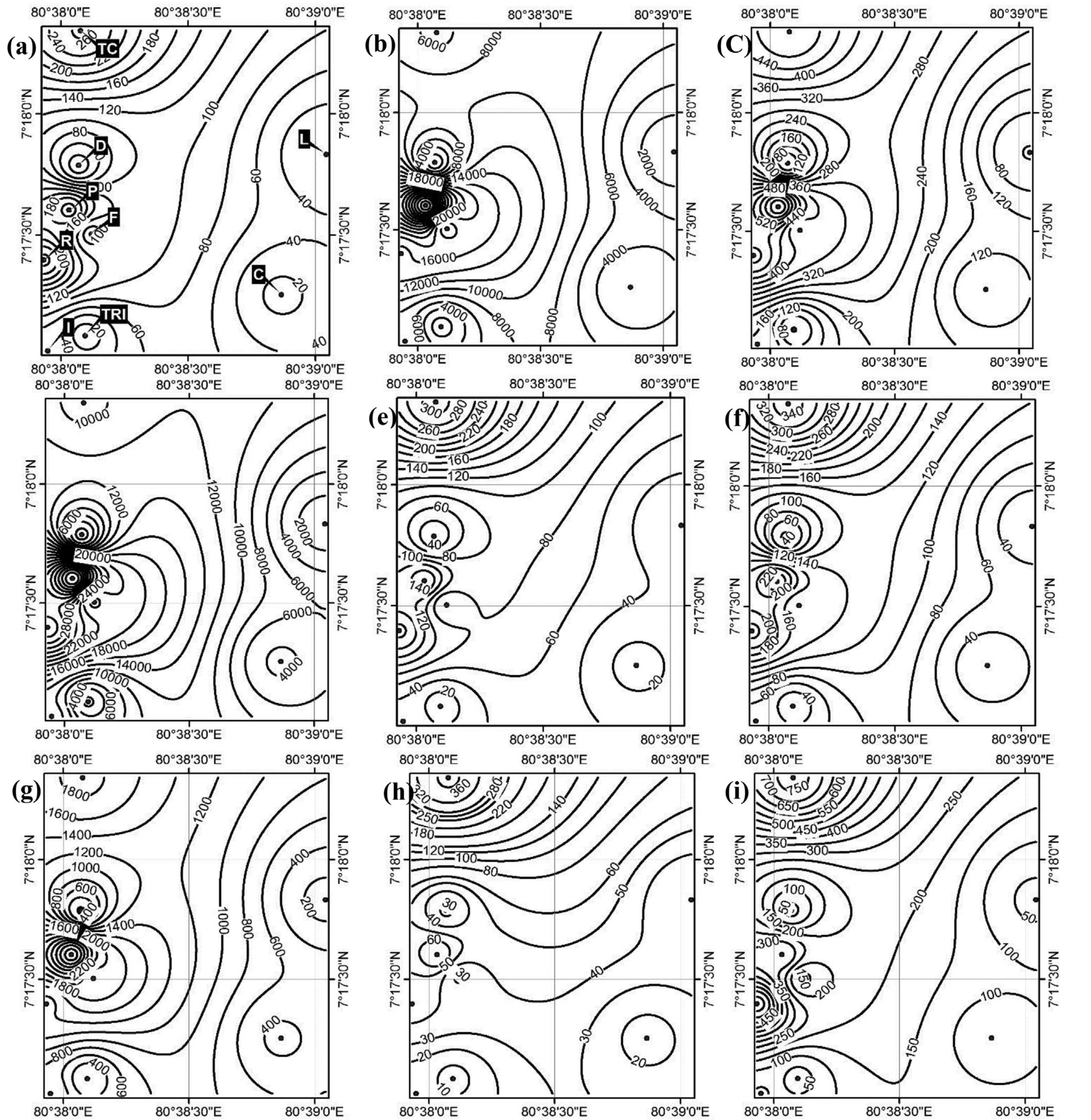


Fig. 2. Spatial distribution of heavy metal at the nine sampling locations in Kandy and its environs. (a) Cr, (b) Al, (c) Mn, (d) Fe, (e) Ni, (f) Cu, (g) Zn, (h) Cd, and (i) Pb; (C) Children's park, (D) Dodanwela, (F) Fire brigade station, (I) National Institute of Fundamental Studies, (L) Lewalla, (P) Police station, (R) Railway station, (TC) Trinity college station, and (TRI) Tea Research Institute (The contours in each map are represent the concentrations of HM loads).

data. The point and raster data were obtained from the world geographic coordinate system of WGS 1984 and the z factor designates the contaminant level and the contours define the concentration and risk ranges. The sample sites C, F, P, R and TC with high traffic volumes were found to have higher metal loads than the other sampling sites. The sample sites identified as having medium traffic activities were also found to have considerable HM concentrations, but lower than those having relatively higher traffic activities. Due to the absence of industrial activities, traffic and other vehicular activities are considered to be the major sources of HMs in Kandy and its environs (Weerasundara et al., 2017).

3.2. Contamination assessment of atmospheric deposition

The CF values for the investigated HMs are shown in Table 1. The data presented show that there is significant enrichment of Pb, Zn, Ni, Cu and Cd. However, Cr and Mn are at low contamination levels, whilst Fe is at the uncontaminated level. These outcomes were further categorized into contamination levels using I_{geo} values, based on which, it can be concluded that Fe is at an uncontaminated

Table 1
Contamination factor (CF) values for heavy metals at different sampling sites.

Site	Metal						
	Cr	Mn	Fe	Ni	Cu	Zn	Pb
C	0.83	0.76	0.09	2.33	2.89	5.38	20.45
D	1.61	0.42	0.03	4.10	4.13	2.64	11.13
F	4.77	3.10	0.48	9.87	16.02	28.33	40.08
I	2.51	1.33	0.26	4.91	5.75	10.46	13.21
L	1.07	0.32	0.02	3.64	2.91	0.02	11.00
P	11.3	5.64	1.07	25.16	26.50	49.25	100.27
R	12.96	4.45	0.81	26.51	25.47	20.11	179.62
TC	13.86	4.31	0.23	49.51	39.35	27.35	221.93
TRI	0.45	0.33	0.04	1.44	2.88	3.07	8.72

level and Pb is at an extremely contaminated level (Fig. 3a).

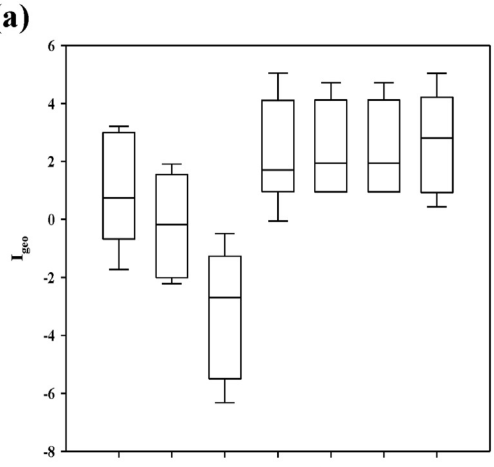
The boxplot for I_{geo} values for atmospheric deposition is presented in Fig. 3a. The mean I_{geo} values were 0.96, −0.15, −0.29, 2.33, 2.51, 2.68 and 4.45 for Cr, Mn, Ni, Cu, Zn and Pb, respectively. The contamination level ranking for the nine sampling sites is shown in Fig. 3b. Comparatively, the maximum I_{geo} values were reported for Pb, Zn, Mn and Cu as shown in Fig. 3a. In the ranking for Pb, all the sampling sites can be categorized into class 3 to 6, indicating that they are in a contaminated state. Interestingly, the sites located in places where the vehicular activities are low, also have moderate contamination levels for Pb. Wind and long-term transportation of dust are attributed to be the reasons for this observation. However, the sites having heavy traffic activities are in the category of extremely contaminated, confirming that contamination is due to vehicular activities. Zn, Mn and Cu are also at the contaminated level in addition to Pb, according to I_{geo} values. Although Fe has high metal concentration in deposition samples, it is in the uncontaminated category, further confirming its crustal origin (Wei et al., 2009; Wei and Yang, 2010). Cr has moderate to heavy contaminated status at the sites with heavy traffic activities. According to the results obtained, the atmosphere in Kandy can be considered as being HM contaminated.

3.3. Health risk assessment

The values of HQ and HI for the nine HMs for the different sampling sites are presented in Table 2 for the three daily dose models. The highest average values of HQ were found to be through the ingestion pathway and decrease in the order of ingestion > dermal contact > inhalation. The contribution of ingestion HQ to HI is the highest among the three exposure pathways. More than 95% of the contribution is through the ingestion pathway for the overall human health risk. Previous studies which investigated atmospheric dust also derived similar results (Du et al., 2013; Lu et al., 2014; Zheng et al., 2010). Therefore, it can be concluded that in the urban environment in Kandy, the ingestion pathway has high possibility to impact on human health. Table 3 shows HI and HQ values reported for a selection of past studies. Compared with HM concentrations in sites from China, where a range of different anthropogenic activities were prominent, such as heavy industrial activities, were much higher than that obtained in this study (Du et al., 2013; Sun et al., 2014; Zahra et al., 2014). Although the data from this study report relatively high HM concentrations, in respect to human health risk, the levels are still within safe limits. According to Table 3, past studies conducted in Angola (Ferreira-Baptista and De Miguel, 2005), Turkey (Kurt-Karakus, 2012), and Australia (Ma et al., 2016), also show similar results. Therefore, though there is environmental pollution, the current impact on human health may not be very severe. A human health risk assessment is generally needed to determine the appropriate risk mitigation strategies.

HQs and HIs for the nine HMs are lower than the safe level for children, indicating that there are no significant health risks from these metals. However, P and R that were categorized as high traffic volume sites show possible health risk from ingestion of Fe (HQs are 1.8 and 1.4, respectively). Atmospheric deposition in Kandy City has high Fe content as shown in Fig. 1. Almost all the sampling sites demonstrated a similar trend. In the ingestion pathway, the average HI ranged as, Fe > Cu > Al > Cd > Ni > Zn > Pb > Cr > Mn. Although CF assessment and I_{geo} assessment showed that Fe has uncontaminated status, Fe is present in atmospheric deposition and can impact on people in the region. Therefore, attention should be given to the health impacts of Fe. As P and R sampling sites are located in the centre of Kandy City, the children who are exposed to the city atmosphere, are at risk due to increased Fe concentrations. Other than Fe, Cu and Al could also exert a significant influence on children's health in the near future as these metals have HI values near to 1 at some of the sampling sites. Through the inhalation

(a)



(b)

Site	Metal						
	Cr	Mn	Fe	Ni	Cu	Zn	Pb
C	0	0	0	1	1	2	4
D	1	0	0	2	2	1	3
F	2	2	0	3	4	5	5
I	1	0	0	2	2	3	4
L	0	0	0	2	1	1	3
P	3	2	0	5	5	6	6
R	4	2	0	5	5	4	6
TC	4	2	0	6	5	5	6
TRI	0	0	0	0	1	2	3

Fig. 3. **(a)** I_{geo} of heavy metals in atmospheric deposition in Kandy. **(b)** Pollution levels at the nine sample collection points. Numbers indicate the 7 classes of pollution classified based on I_{geo}. 0 - Practically uncontaminated, 1 – uncontaminated to moderately contaminated, 2 – Moderately contaminated, 3 – Moderately contaminated to heavily contaminated, 4 – Heavily contaminated, 5 – Heavily contaminated to extremely contaminated, 6 – Extremely contaminated.

Table 2
Average Hazard Quotient (HQ) and Hazard Index (HI) values for nine heavy metals based on three daily dose models via ingestion pathway, dermal contact pathway and inhalation pathway.

Metal	Concentration mg/kg		HQ_{ing} (10^{-4})		HQ_{inh} (10^{-4})		HQ_{dermal} (10^{-4})		HI (10^{-4})	
			Children	Adult	Children	Adult	Children	Adult	Children	Adult
Al	Min	507.2	284	6	0.00122	0.00103	0.538	0.347	65	7
	Max	43327.6	5539	539	0.104	0.0879	49	29	5589	623
	Mean	10119.7	1193	138	0.0245	0.0205	11	6	1305	145
Cr	Min	8.5	3	1	0.0000001	0.0000005	0.00007	0.000183	3	1
	Max	261.8	95	40	0.000004	0.000017	0.00237	0.00564	95	41
	Mean	103.0	37	16	0.000001	0.000006	0.00932	0.00222	37	16
Mn	Min	38.5	0.3	0.008	0.311	0.0894	0.00493	0.00293	0.624	0.101
	Max	672.1	5	0.144	5	1	0.0859	0.0511	11	1
	Mean	273.6	2	0.05	2	0.635	0.035	0.0208	4	0.714
Fe	Min	770.8	328	35	0.000474	0.00223	1	0.754	329	35
	Max	43922.5	18718	2005	0.0122	0.127	72	42	18791	2048
	Mean	13774.9	5870	628	0.00508	0.0399	22	13	5893	642
Ni	Min	8.9	56	6	0.00003	0.0000254	1	0.609	57	6
	Max	306.4	1959	209	0.00105	0.00876	35	20	1994	230
	Mean	87.6	560	60	0.000299	0.000251	10	6	570	66
Cu	Min	25.5	651	69	0.0123	0.0103	5	3	657	73
	Max	347.7	8892	952	0.168	0.141	80	47	8972	1000
	Mean	123.6	3161	338	0.0599	0.0502	28	16	3190	355
Zn	Min	136.9	58	6	0.00101	0.000842	0.478	0.284	58	6
	Max	3331.4	1419	152	0.0244	0.0205	11	6	1431	159
	Mean	1116.9	476	51	0.0082	0.00687	3	2	479	53
Cd	Min	6.5	70	30	0.0000001	0.0000004	0.00006	0.000155	70	30
	Max	386.5	4235	1815	0.0000081	0.00027	0.00389	0.00926	4235	1815
	Mean	68.8	754	323	0.0000014	0.000004	0.000693	0.00165	754	323
Pb	Min	30.3	36	3	0.0000042	0.000014	0.00199	0.00475	36	3
	Max	772.1	940	101	0.000107	0.00035	0.0508	0.121	940	100
	Mean	234.4	285	30	0.000032	0.000109	0.0154	0.0367	285	30

pathway, the HI values decrease in the order of Mn > Cu > Al > Zn > Fe > Ni > Pb > Cr > Cd. However, all the HI values are far less than 1. Therefore, the health risk through inhalation pathway is not significant in Kandy and its environs. Similarly, the dermal HI values are lower than 1 and there is no significant health risk through dermal contact of atmospheric HM. HI_{dermal} decreases as Cu > Fe > Al > Ni > Zn > Mn > Pb > Cr > Cd.

For adults, HQ and HI values are below 1, which suggests that there are no significant adverse health impacts on adults due to HMs in atmospheric deposition. The changing pattern of HI was observed to be similar to that for children. For adults, the highest HI for the ingestion pathway was recorded for Fe (HI = 0.2) and it is also much lower than the threshold safe level. For the inhalation pathway, the highest HI was recorded for Mn (HI = 0.0001) and for dermal contact the highest HI was for Cu (HI = 0.004) and these values are also lower than the threshold levels. Therefore, the potential health risk to adults due to atmospheric pollution of HMs is not significant. Compared to children, the health risk for adults is much lower.

Considering carcinogenic health impact, the HI for carcinogenic metals also have lower HI values (HI < 1) indicating that there is no significant cancer risk due to HMs in atmospheric deposition in Kandy. The lifetime average cancer risk also falls within the threshold range ($10^{-6} - 10^{-4}$) or below. This means that currently there is no cancer risk to the population in Kandy City and its environs in relation to the nine heavy metals investigated in the atmospheric deposition.

However, past studies have shown that on a yearly basis, particulate matter emissions from traffic sources have an increasing trend in the Kandy area (Seneviratne et al., 2017). In a study conducted over 2 years from 2012 to 2014, traffic based particulate matter concentrations were found to have increased considerably. In 2012, particulate matter concentrations were in the average range of 0.0–0.5 μg/m^3, and by 2014 it had increased to 0.75–1.7 μg/m^3 (Seneviratne et al., 2017). Therefore, appropriate measures are needed to mitigate the currently increasing pollution trends in order to prevent serious health issues into the future.

Moreover, it is important to note that with this increasing trend of atmospheric pollution, the entire population may not face the risk in a similar manner as the lifestyle and the occupation has a significant influence in this regard. Considering the residents in the Kandy area who reside near roadways and spend most of the day at home such as the elderly and little children, and street vendors would be at a higher risk from heavy metals in the atmosphere due to their long exposure periods (Wickramasinghe et al., 2012). School children are the next population group that would be at considerable risk from atmospheric heavy metals as they are exposed to polluted atmosphere during peak hours in the morning and midday. Also, many schools in Kandy City are located near roads with heavy traffic congestion. Working population in Kandy City also having great deal with atmospheric heavy metal deposition but not much as lifetime city personals as most of their exposure time limit to more or less 8 h.

4. Conclusions

Al and Fe, from both geogenic and anthropogenic sources, were found to be in significantly higher concentrations in atmospheric deposition compared to the other seven metals investigated. Zn also had elevated concentrations in the atmosphere. All the other metals investigated, namely, Cr, Mn, Ni, Cu, Pb and Cd were reported in lower concentrations in the atmospheric deposition in the study area. Sample sites, which were categorized as having high traffic activities, exhibited increased heavy metal concentrations in deposition samples. Contamination Factor (CF) values and I_{geo} values confirmed that Al and Fe are in the 'no contamination' range. The major exposure pathway of heavy metals for both, children and adults is ingestion. The hazard quotient for the pathways investigated in this study was in the order of ingestion > dermal contact > inhalation. Both, hazard quotient and hazard index were less

Table 3
Comparison of Hazard Quotient (HQ) and Hazard Index (HI) values for heavy metals reported in different studies.

Metal	Location	$HQ_{ing}(10^{-4})$		$HQ_{inh}(10^{-4})$		$HQ_{dermal}(10^{-4})$		HI (10^{-4})		Reference
		Children	Adult	Children	Adult	Children	Adult	Children	Adult	
Al	Kandy, Sri Lanka	1293	138	0.0245	0.0205	11	6	1305	145	This study
Cr		37	16	0.000196	0.000006	0.00932	0.0022	37	16	
Mn		2	0.05	2	0.635	0.035	0.0208	4	0.714	
Fe		5870	628	0.00508	0.0399	22	13	5893	642	
Ni		560	60	0.000299	0.000251	10	6	570	66	
Cu		3161	338	0.0599	0.0502	28	16	3190	355	
Zn		476	51	0.0082	0.00687	3	2	479	53	
Cd		754	323	0.000001	0.000004	0.000693	0.00165	754	323	
Pb		285	30	0.00003	0.0001	0.0154	0.0367	285	30	
Cr	Beijing, China	7720	23.8	5.87	6.29	319	238	8050	268	Du et al. (2013)
Ni		166	22.3	0.00305	0.00327	29.9	22.3	196	44.6	
Cu		249	33.5	0.00435	0.00465	43.7	23.6	293	57.1	
Zn		9330	12.5	0.00177	0.0019	4.20	3.13	97.5	15.6	
Cd		81.8	11	0.00155	0.00116	14.7	11	96.5	22	
Pb		7370	989	0.139	0.149	42	330	7410	1320	
Cr	Nanjing, China	2460	345	590	590					Sun et al. (2014)
Ni		70.1	981	615	615					
Cu		5250	736							
Mn		324	45.4	7040	7040					
Zn		1440	202							
Cd		3390	470	2840	2840					
Pb		57600	8070							
Cr	Taiyuan, China	8300	1100	28	30	1500	1100			Zhang et al. (2016)
Ni		510	69	0.0095	0.01	93	69			
Cu		470	62	0.0081	0.0089	82	61			
Mn		2600	350	160	180	460	340			
Zn		570	77	0.011	0.012	26	19			
Cd		1300	170	0.024	0.026	230	170			
Pb		23000	3100	0.44	0.49	1400	1000			
Al	Luanda, Angola	333		6.5		9.32		349		Ferreira-Baptista and De Miguel (2005)
Cr		580		1.7		81.2		663		
Ni		35.2		0.00098		0.365		35.5		
Cu		0.0072		0.00202		0.675		73		
Mn		386		34.7		27		448		
Zn		73		0.00204		1.02		74		
Cd		79.1		0.0022		22.1		101		
Pb		7100		0.198		132		7320		
Cr	Istanbul, Turkey	310	58	0.0029	0.0028	25	180			Kurt-Karakus (2012)
Mn		130	26	0.0012	0.0013	10.3	82			
Ni		1200	180	0.011	0.0089	106	58			
Cu		1200	130	0.011	0.0064	91.2	420			
Zn		380	49	0.0035	0.0024	29	160			
Cd		350	3.4	0.0033	0.0017	27.7	110			
Pb		1500	230	0.014	0.011	119	740			
Al	Gold Coast, Australia							672		Ma et al. (2016)
Cr	(Average values)							2030		
Mn								1120		
Fe								523		
Ni								30		
Cu								155		
Zn								45.5		
Cd								169		
Pb								1110		

than 1. The lifetime daily cancer risk also ranged within the acceptable limit, indicating that there is no immediate possibility of cancer risk due to the presence of heavy metals in the atmosphere, but this could change in the long-term with increased air pollution in Kandy, unless appropriate mitigation measures are implemented. Considering the patterns of heavy metal loads within Kandy City, it can be concluded that mitigation strategies such as easing of traffic congestion, providing bypass arrangements and improving fuel and vehicle quality, and phasing out the use of Zn coated roofing materials will be able to change the current situation in a positive way. Hence, the study findings are of value to regulatory authorities for the implementation of appropriate strategies to alleviate heavy metal pollution in the Kandy City environment. The study clearly identifies the locations with high pollution levels and the risk levels in the City.

Acknowledgment

The Authors wish to offer a special acknowledgment to National Science Foundation (NSF), Sri Lanka, for providing funds for undertaking this research study (Grant number RG/2014/EB/03).

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jenvman.2018.04.036.

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