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TRACE ELEMENT CHARACTERIZATION IN HOUSEHOLD DUSTS IN INDUSTRIAL AREAS ALONG HIGHWAYS IN BANGLADESH AND THEIR HEALTH IMPLICATIONS

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ABSTRACT

A study was carried out in the industrial areas in order to determine the composition of trace elements in dusts deposited at homes, identifying different sources of elements and also for their possible health impacts. A total of 14 deposited dust samples were collected from households near industrial areas of Tangail and Gazipur districts during sampling campaign. Samples were analyzed using X-ray Fluorescence and the highest concentration was observed for Fe whereas the overall rank of the concentrations is Fe>Ca>K>Ti>Zn>Pb>Zr>Sr>Rb>Cu>Y>As>Cr>Ni. The Correlation analysis, enrichment factor and factor analysis indicated that anthropogenic sources especially industrial activity, vehicular emission and household cooking are the main sources of trace elements in the study areas. The non-carcinogenic and carcinogenic health risks indicated that children are more vulnerable for non-carcinogenic effects whereas the values of cancer risk for both the child and adult are below the acceptable limit of European Union.

Keywords: Indoor air, trace elements, industrial community, health risk, sources, factor analysis

INTRODUCTION

Bangladesh is an agricultural country; recently economic growth has improved the quality of life for most of the population, but it progressively degrades the environmental quality. The quality of air, water and soil is deteriorated due to different industrial activities such as garments and textile industries, pulp and paper, pharmaceuticals, metal processing, food industry, fertilizers, glass industry, cement industry, dyeing and painting etc. As a result, people living near the industrial areas are more vulnerable to heavy metal pollution. There is an increasing concern about heavy metals contamination in indoor environment as they can enter deep into the lungs and result in serious health effects (Kura *et al.* 2013). The composition of inhalable particulate matter ($D_{aerodynamic\ diameter}$ ≤100 µm; they enter the respiratory tract including head airways) is complex and differs depending on the source and location. The occurrence of toxic metals such as Pb, Zn, Cu, and Ni in inhalable particulates may contribute to substantial health effects (Safo-Adu *et al.* 2014). Some of these heavy metals are carcinogenic and mutagenic.

Dust is an important medium that can give information about the level, distribution and fate of contaminants present in the environment. Dust consists of solid matter or particulate in the form of fine powder (less 100 µm), lying on the ground or on the surface of objects or blown about by natural forces or mechanical forces. As the composition of settled dust is similar to atmospheric suspended particulates, it can be also an indicator of pollutants such as heavy metal contamination in the atmosphere (Leung *et al.* 2008). Indoor dust may contain toxic materials, particularly heavy metals, in quantities that may be potentially harmful to human health (Aucott and Caldarelli 2012). Indoor dust has been found to contain a quantity

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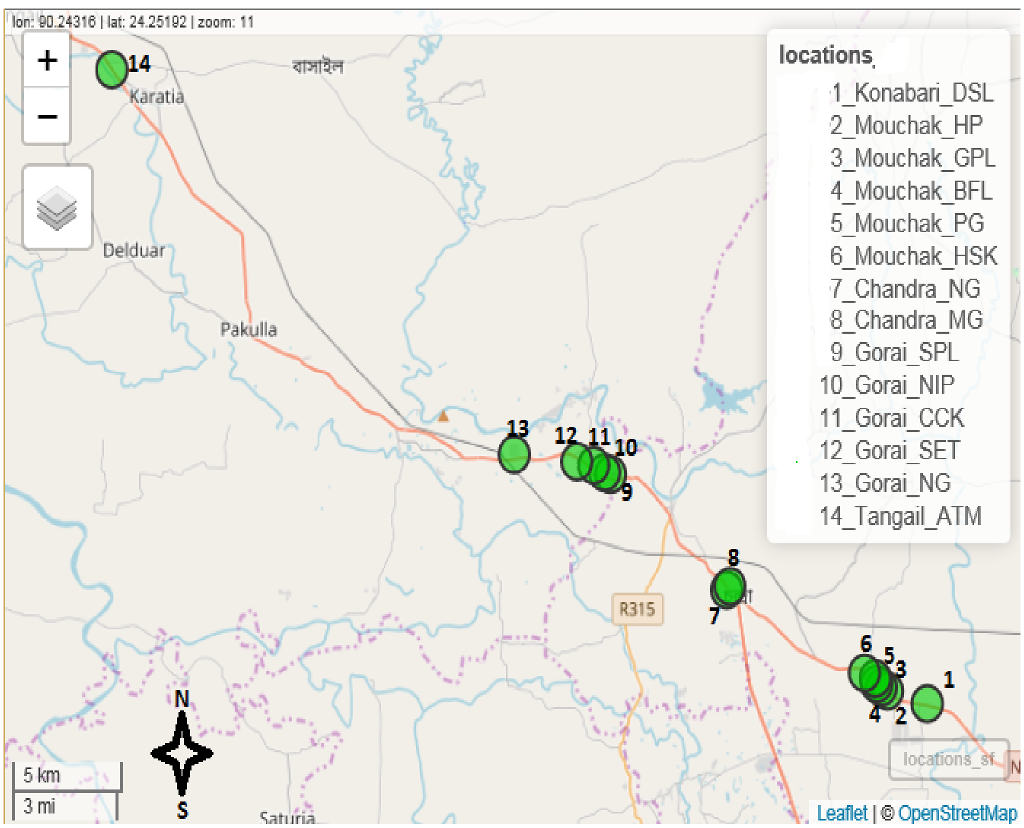
of heavy metals which may potentially affect the health of young children (Latif *et al.* 2009). Humans can become exposed to heavy metals in dust through several routes which include ingestion, inhalation, and dermal absorption (Leung *et al*. 2008).

Therefore, studying on the characteristics dust pollution is not only an important aspect of evaluation of quality of urban environment, but also of great significance for human health. Several researches have been carried out to characterize elemental composition of road dust (Rakib *et al.* 2014: Dhaka; Ahmed and Ishiga, 2006: Dhaka) but research related to characterization of household dust is limited. Therefore, this research was conducted to fulfill the following objectives (i) to determine the concentration of trace elements in dust deposited at homes near industrial zones, (ii) to assess the possible health risks on residents and (iii) to identify the sources of elements present in indoor air deposited at homes.

MATERIALS AND METHODS

Study Area

In this study, fourteen samples were collected from households located near the industrial areas. The features of the sampling sites are given in Table 1 (Map 1). Dust samples were collected into 40 mL test tube bottles by gently sweeping from different surfaces. The dust samples were screened to remove any visible hair, soil, and grit.



Map 1. Sampling locations of the study area (Details of the locations 1 to 14 are given in Table 1)

Table 1. Key features of the sampling locations of households located near the industrial areas

Sample No.	Name of Industry	Types of Industry	Place	Distance from Industry	Latitude (Decimal Degrees)	Longitude (Decimal Degrees)
1	Designtext Sweater Ltd.	Chemicals	Konabari, Gazipur	300 m.	24.011099	90.323845
2	HUSDON pharma	Chemicals	Mouchak, Gazipur	250 ft.	24.015906	90.306378
3	General Pharmaceuticals Ltd.	Chemicals	Mouchak, Kaliakur Gazipur	100 ft.	24.017265	90.303375
4	Bay Footwear Limited & Agro Industry	Chemicals	Mouchak, Kaliakur, Gazipur	350 ft.	24.018505	90.30175
5	Purbani Groups (Karim Textile)	Dyeing(Chemicals), Garments	Mouchak, Kaliakur, Gazipur	320 ft.	24.019977	90.300454
6	Hydroxite & Syntex Knit Wear Limited	Garments	Mouchak, Kaliakur, Gazipur	275 ft.	24.022294	90.295933
7	Nur Group	Dyeing	Chandra, Gazipur	200 ft.	24.050919	90.23444
8	Mahmud Group	Dyeing & Washing	Chandra, Gazipur	300 ft.	24.051867	90.23545
9	SQUARE Pharmaceuticals Ltd.	Chemicals	Gorai, Mirzapur, Tangail	250 ft.	24.090947	90.181849
10	Necuage Industrial Pearls	Garments, Dyeing(Chemical)	Gorai , Mirzapur, Tangail	300 ft.	24.91463	90.179007
11	Comfit Composite Knit Limited	T-Shirt Dyeing , Have EPP	Gorai, Mirzapur, Tangail	100 ft.	24.094385	90.174686
12	South East Texttile (pvt.) Ltd.	Textiles	Gorai, Mirzapur, Tangail	150 ft.	24.095072	90.16713
13	Nasir Glassware Tube Industries Ltd	Glass & Tube, Chemicals	Gorai, Mirzapur, Tangail	300 ft.	24.097934	90.138715
14	Alauddin Textile Mills	Textiles, Dyeing,Chemicals	Tangail	100 ft.	24.23112	89.957951

Sample Preparation and Analysis

Samples were dried in an oven at 70^{o}C for overnight in order to free moisture and bottled in clean plastic bottles. Then the samples were analyzed using Energy Dispersive X-ray Fluorescence (EDXRF) at the laboratory of Chemistry division, Bangladesh Atomic Energy Centre, Dhaka, Bangladesh. In the XRF measurements, the samples were processed into 1.0 cm diameter and 1 mm thick pellets of weight approximately 0.1g. Similar procedures were followed for the preparation of standards from IAEA-Soil 7, Montana-1. Here, IAEA-Soil 7 was used as a standard and Montana-1 was used as the quality assurance and quality control (QA/QC) in the measurement process.

The minimum detection limit (MDL) of the method has been determined with Montana-1 standard for 1000 sec irradiation time.

Health risk Assessment

Elements entered into human body through three exposure pathways: (a) ingestion, (b) dermal contact, and (c) inhalation (Ferreira-Baptista and Miguel 2005; Kurt-Karakus 2012). The potential exposure doses through ingestion, inhalation and dermal contact were calculated from the following equations (*Eq.*) as recommended by USEPA (1989). The exposure parameters used for the calculation of health risk assessment are given in Table 2.

Daily potential exposure dose through ingestion

$$ADI_{ing} = \frac{C \times IR \times EF \times ED \times CF}{BW \times AT} \quad (Eq. 1)$$

Daily potential exposure dose through inhalation

$$ADI_{inh} = \frac{C \times IR_{air} \times EF \times ED}{BW \times AT \times PEF} \quad (Eq. 2)$$

Daily potential exposure dose through dermal contact

$$ADI_{der} = \frac{C \times SA \times FE \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (Eq. 3)$$

Finally, non-carcinogenic and carcinogenic risk were calculated from the term described by USEPA and is given in *Eq. 4 and Eq. 5.*

Non-carcinogenic Risk Assessment

Hazard quotient (HQ) = $\frac{ADI}{RfD}$ *(Eq. 4)*

where ADI= Exposure Dose

RfD= Chronic Reference Dose in mg/kg-day of a heavy metal

Carcinogenic Risk Assessment

Cancer Risk $(CR) = ADI_k \times CSF_k$ *(Eq. 5)*

where ADI_k = The average daily intake for kth trace element

CSF_k = Cancer slope factor for n number of trace element

Table 2. Recommended values of the parameters used to calculate the daily exposure dose of trace elements in Dust

Parameter	Definition	Child	Adult	References
C	Average concentration of HMs in APM (mg/kg)			Hu *et al.* (2012)
BW	Body Weight (kg)	15	70	USEPA(2004)
EF	Exposure frequency (days/year)	350	350	DoEA (2010)
ED	Exposure duration (year)	6	30	DoEA (2010)
IR	Ingestion rate (mg/day)	200	100	DoEA (2010)
IRair	Inhalation rate (m^3/day)	10	20	DoEA (2010)
SA	Skin surface area (cm^2)	2100	5800	DoEA (2010)
AF	Soil adherence factor (mg/cm^2)	0.2	0.07	DoEA (2010)
ABS	Dermal Absorption factor	0.1	0.1	DoEA (2010)
FE	Dermal Exposure ration	0.61	0.61	DoEA (2010)
PEF	Particulate emission factor (m^3/kg)	1.3×10^9	1.3×10^9	DoEA (2010)
CF	Conversion factor (kg/mg)	10^{-6}	10^{-6}	DoEA (2010)
AT	Averaging Time: For carcinogens (days)	365×70	365×70	DoEA (2010)
	For non-carcinogens (days)	365×ED	365×ED	DoEA (2010)

RESULTS AND DISCUSSION

Characterization of indoor air deposited at homes in industrial community

The mean concentration of potassium (K) was 10892 mg kg^{-1} and ranged from 4668 mg kg^{-1} to14665 mg kg^{-1}. The higher concentration of K was observed in samples S14, S13, S12 and S2, whereas the lower concentration was found in samples S1 and S9 (Table 3). In this study, the higher concentration of potassium was found in several locations especially near Alauddin Textile mill (S14), Tangail. Biomass burning in household activities may be the main reason of the availability of K in that area. The concentration of Calcium (Ca) ranged from 14280 mg kg^{-1} to 44845 mg kg^{-1} whereas the mean value of Ca concentration is 30898 mg kg^{-1}. The higher concentration of calcium was observed in several locations especially near SQUARE Pharmaceuticals Ltd. which is located in Mirzapur, Tangail. The observed mean concentration of Ti was 4349 mg kg^{-1} and varied between 2753 mg kg^{-1} to 5401 mg kg^{-1}. The higher concentration of Ti was observed in S13, S14, S5 and S6 samples whereas the lower concentrations were found in samples S1 and S5 (Table 3). The average Fe concentration was 57435 mg kg^{-1} and fluctuated from 36080 mg kg^{-1} to 171780 mg kg^{-1}. The highest Fe concentration was observed in S3 whereas the lower Fe concentration was observed in all remaining samples except S4 (Table 3). The highest concentration of Fe was found in Designtext Sweater Ltd that is situated in Konabari, Gazipur. Natural source was considered as main source of iron in this region. Fe is an abundant element in the earth crust (Al-Khashman 2004). The concentration of Cu ranged from 16 mg kg^{-1} to 41 mg kg^{-1} and its mean value was 25 mg kg^{-1}. However the higher Cu concentration was found in samples S2 and S14 whereas the lower concentration was detected in samples S5, S11 and S13 (Table 3). Street dust was the major source of Cu. Cu is usually emitted in the environment through combustion of fuel. The observed mean value of Zinc concentration was 1718 mg kg^{-1} and it varied from 561 mg kg^{-1} to 5863 mg kg^{-1}. The highest Zinc concentration was found in sample S2 whereas the lower Zinc concentration was observed in samples S5, S8, S12, S13 and S14 (Table 3). The accumulation of Zn emitted from tyres, motor oil and the usage of motor vehicle brakes (Han *et al.* 2011). The average value of As concentration was 9 mg kg^{-1} which ranged from 6 mg kg^{-1} to 12 mg kg^{-1}. On the basis of result, higher concentration of As was found in the samples S2, S6, S7 and S9 whereas the lower concentration of As was observed in S1, S5, S13 and S14 (Table 3). Natural source was considered as main source of As contamination in the earth. Comparing with different locations, the observed mean value of Rb concentration was 126 mg kg^{-1} and it ranged from 71 mg kg^{-1} to 284 mg kg^{-1}. The highest Rb concentration was found in sample S14 whereas the lower Rb concentration was found in samples S1, S3, S9 and S10. In this study the highest Rb ion concentration was observed near Alauddin Textile Mills at Tangail. Data studied represent mean value of strontium (Sr) concentration was 155 mg kg^{-1} and it ranged from 106 mg kg^{-1} to 202 mg kg^{-1}. In this study the highest Sr ion concentration was observed in several locations especially near Bay Footwear Limited & Agro Industry at Mouchak, Kaliakur, Gazipur. The average concentration of Ytrium (Y) was 24 mg kg^{-1} and it ranged from 15 mg kg^{-1}-37 mg kg^{-1}. Higher Y concentration was observed in several locations especially near General Pharmaceuticals Ltd at Kaliakur, Gazipur. Y may originate from paint along with gaps and cracks in walls and building materials (Tong and Lam 2000). Mean Zirconium (Zr) concentration was 238 mg kg^{-1} and it ranged from 142 to 474 mg kg^{-1}. The highest Zr concentration was observed in several locations especially near General Pharmaceuticals Ltd. at Kaliakur, Gazipur. Zr was identified as anthropogenic sources in road dust (Atiemo *et al.* 2011). The observed mean concentration of Pb was 310 mg kg^{-1} and it varied from 202 mg kg^{-1} to 785 mg kg^{-1}. In this study the highest concentration of lead was observed in near Alauddin Textile Mills, at Tangail. Pb is also frequently used in different industries such as dyes and paints.

Table 3. Concentration of trace elements present in household dust ($mgkg^{-1}$)

Sample	K	Ca	Ti	Cr	Fe	Ni	Cu	Zn	As	Rb	Sr	Y	Zr	Pb
S1	4668	18555	2753	<5.2	36080	<0.19	23	2171	6	86	125	23	187	233
S2	13230	35455	4256	<5.2	45095	<0.19	37	5863	7	120	176	23	177	417
S3	10154	33115	4290	<5.2	171780	<0.19	30	1707	7	71	106	17	159	225
S4	10410	33560	4179	<5.2	75285	<0.19	29	2790	6	116	191	25	250	273
S5	8396	14280	4972	<5.2	38265	<0.19	20	667	6	144	108	37	474	202
S6	12660	36840	4860	<5.2	49705	<0.19	22	1180	7	134	202	36	365	219
S7	11990	41205	4070	<5.2	47680	<0.19	22	1209	6	123	180	30	285	213
S8	11002	34445	4779	<5.2	44130	<0.19	22	765	10	135	173	28	269	326
S9	7856	44845	2764	<5.2	50785	<0.19	27	1324	10	82	130	16	164	280
S10	9442	34570	3969	<5.2	41885	<0.19	22	2194	12	106	156	26	243	378
S11	10005	37405	4865	<5.2	48100	<0.19	18	2208	12	116	166	26	217	289
S12	13840	28510	4672	<5.2	47225	<0.19	20	561	11	122	166	16	142	248
S13	14165	19230	5401	<5.2	55700	<0.19	16	558	12	129	121	15	153	259
S14	14665	20550	5051	<5.2	52370	<0.19	41	853	12	284	163	24	242	785
Mean	10892	30898	4349		57435		24.9	1717.9	8.9	126.3	154.5	24.4	237.6	310.5

Sources of Elements Present in Indoor Air Deposited at Homes

A Pearson correlation was conducted to observe the relationship among metals. The correlation coefficient of the analysis is given in Table 4. Strong correlation was observed between pairs of Y-Zr (r=0.93), Pb-Rb(r=0.82), K-Ti (r=0.73) and Cu-Pb (r=0.73). Statistically significant relationship was also observed between pairs of K-Rb, Ti-Rb and Ca-Sr. This significant relationship indicated that these metals may be emitted from the same sources.

Correlations among elements

Table 4. A correlation matrix between different elements

	K	Ca	Ti	Fe	Cu	Zn	As	Rb	Sr	Y	Zr	Pb
K	1.00											
Ca	0.03	1.00										
Ti	0.73*	-0.32	1.00									
Fe	0.25	0.22	0.13	1.00								
Cu	0.23	0.06	-0.13	0.29	1.00							
Zn	-0.06	0.28	-0.28	0.04	0.47	1.00						
As	0.36	0.00	0.32	-0.11	-0.12	-0.29	1.00					
Rb	0.56	-0.39	0.53	-0.08	0.46	-0.24	0.33	1.00				
Sr	0.44	0.50	0.18	0.06	0.15	0.25	-0.02	0.25	1.00			
Y	-0.11	-0.05	0.24	-0.35	-0.13	-0.04	-0.42	0.23	0.36	1.00		
Zr	0.12	-0.21	0.31	-0.28	-0.17	-0.24	-0.39	0.27	0.13	0.93*	1.00	
Pb	0.42	-0.17	0.20	-0.02	0.73*	0.12	0.46	0.82*	0.19	-0.09	-0.12	1.00

- Data with star sign (*) showed statistically significant relationship.

Enrichment Factors

Enrichment factors indicate the source of a metal in a particular place. Usually, the enrichment of metals in atmospheric particles relativel to the upper surface dust composition is an indication of emissions from anthropogenic sources. The enrichment factor (EF) for the metals was calculated using the following equation:

$$EF = \frac{[E]atm / [R]atm}{[E]soil / [R]soil}$$

Where $[E]_{atm}$ and $[R]_{atm}$ are the mean concentrations of the metals and the reference metal in the atmosphere, and $[E]_{soil}$ and $[R]_{soil}$ are the mean concentrations of the metals and the reference metal in the Earth's crust, respectively. An element is considered to be crustal origin if EF<1, while the element is supposed to be non-crustal origin for EF value greater than 5 (Gao *et al.* 2002). Usually Al, Fe, Si and Ti are given priorities as reference materials because of lacking of predominant anthropogenic sources (Petaloti *et al.* 2006). In this study, Fe is selected as reference material (Alves *et al.* 2015) and the calculated EFs of metals are given in Fig. 1. Considering all sites, the most enriched elements (EF>5) were Zn, Pb and As suggesting that these elements are mainly coming to the atmosphere from the emissions of various anthropogenic activities. Major sources of these elements are considered to be automotive emissions and biomass burning. Low EF values (<5) were found for K, Ca, Ti, Cu, Rb, Sr, Y, Zr at all sampling sites revealing that these elements may originate mainly from soil or road dust re-suspension. In general, metals that are distributed mostly in fine particles show high EF values whereas elements linked with coarse particles give lower EF values.

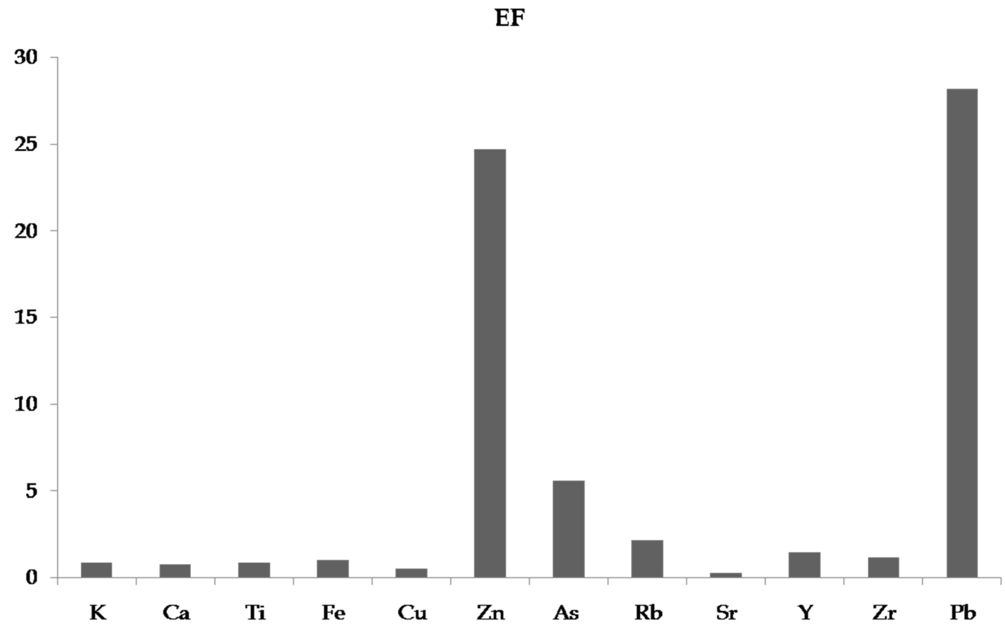


Fig. 1. Enrichment factors (EF) of the elements collected from households

Factor Analysis

To quantify possible sources of household dust, a statistical procedure named Factor Analysis (FA) was performed to identify number of factors and species profile of each source. Main principle of the FA is to reduce the number variables keeping original information. Factors were identified using varimax rotation method based on eigen-value, scree plot, variability in the number of factors and sensibility of each variable to factor loading. Variables were considered as sources when factor loading were >0.75. Potential sources of household dust were identified and are given in Table 5. The first factor includes Cu and Pb. The chemical Cu and Pb are the predominant sources of vehicular emission (Cheng *et al*. 2010). Therefore this factor is designed as vehicular emission. Dust from brake/tire wear contained Cu and Pb. Factor 2 comprised of Y and Zr. Yttrium (Y) and Zr are considered as both natural and anthropogenic oriented chemicals. So this has beer denoted as mixed source. However, the other sources of Zr are heat-shock, furnace linings, foundry bricks, abrasives glass and ceramics industries, scissors and knives, cosmetics, antiperspirants, food packaging and microwave filters. The most common uses of yttrium are LEDs and phosphate, especially the red phosphors in television set. In Factor 3, the highest loading was observed in Ca and Sr. Construction industry such as cement may be the possible source of Ca and Sr. The 4th Factor concludes K and Ti. Biomass burning in household's actives may be the possible source of K in that area. On the contrary, Ti was recognized as anthropogenic source such as road dust, natural crustal, vehicular emissions, and corroded vehicular parts (Atiemo *et al.* 2011).

Table 5. Variables with loading in factor analysis at Tangail and Gazipur. (Variables with loading factors >0.75 are bold marked)

Chemical	Factor 1	Factor 2	Factor 3	Factor 4
K	0.28	0.11	0.33	**0.81**
Ca	-0.18	0.17	**0.81**	-0.15
Ti	0.02	-0.23	-0.02	**0.86**
Fe	0.03	0.39	0.42	0.12
Cu	**0.91**	0.12	0.20	-0.16
Zn	0.41	0.07	0.48	-0.50
As	0.06	0.49	-0.21	0.62
Rb	0.68	-0.26	-0.20	0.61
Sr	0.17	-0.30	**0.78**	0.29
Y	-0.02	**-0.98**	0.09	0.02
Zr	-0.08	-0.95	-0.11	0.09
Pb	**0.90**	0.10	-0.10	0.33
% Variance	2.44	2.49	1.78	2.62
Cumulative Variance	20	21	15	22
Sources	Vehicular emission	Mixed Sources	Construction industry	Household combustion

Non-carcinogenic and carcinogenic health risk

Non-carcinogenic risk for an element is expressed by the term called hazard quotient (HQ) whereas the non-carcinogenic effect to the population is calculated by the term Hazard Index that is the result of the summation of all the HQs due to individual element. People are risked for possible non-carcinogenic effects, if HQ and HI values exceed 1. For a child, the HQ values are higher than 1 for ingestion pathways mainly due to Pb whereas it is less than 1 for both inhalation and dermal pathways (Fig. 2a). For adult, the calculated HI value is less than 1, indicating that the exposured population are not risk for non-carcinogenic effects. The cancer risk of Pb and As has been estimated from the average daily exposure doses for both child and adult population (Fig. 2b). The values of cancer risk for both the trace elements are below the acceptable limit of European Union (10^{-6} to 10^{-4} per year: EC 2001), indicated that these two elements are not responsible for carcinogenic risk.

Fig. 2. Non-carcinogenic and carcinogenic risk due to the exposure of trace elements

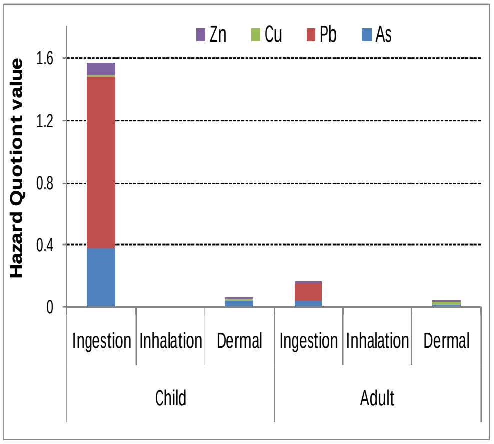


Fig. 2a. Hazard Quotient (HQ) values of trace element for child and adult

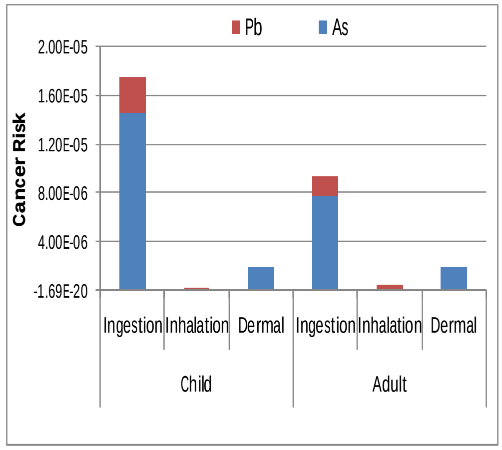


Fig. 2b. Cancer risk values of trace elements for child and adult

CONCLUSION

Trace element concentration at homes in industrial-zones may vary from place to place depending on pollutant types and accumulation and/or depositional factor. The highest concentration was observed for Fe whereas the overall rank of the concentrations is Fe>Ca>K>Ti>Zn>Pb>Zr>Sr>Rb>Cu>Y>As>Cr >Ni. Generally, trace element concentrations in indoor dust are in the range of the corresponding values reported in literature for a variety of locations. Correlation analysis, enrichment factors and factor analysis were performed to identify the possible sources of indoor dust. Strong correlation was observed between pairs of Y-Zr (r = 0.93), Pb-Rb (r = 0.82), K-Ti (r = 0.73) and Cu-Pb (r = 0.73). Statistically significant relationship was also observed between pairs of K-Rb, Ti-Rb and Ca-Sr. This significant relationship indicates that these metals may originate from the same sources. Enrichment Factors might be attributed to different anthropogenic sources of the metals in the indoor environments.

From the results it could be concluded that the concentration of different chemicals in the study areas may have originated from the earlier industrial activities and also vehicular emissions. Possible sources of metals were identified by the statistical procedure named factor analysis and it indicated that main source of Cu and Pb were from vehicular emission whereas main source of Ca and Sr were from construction works or from cement industry. The non-carcinogenic and carcinogenic health risks were calculated and indicated that children are more vulnerable for non-carcinogenic effects whereas the values of cancer risk for both the child and adult are below the acceptable limit of European Union (10^{-6} to 10^{-4} per year).

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