Düzce Üniversitesi
Journal of Science & Technology

Research Article

Estimation of Health Risks Associated with Household Dust Contamination in Bolu (Turkey)

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DOI: 10.29130/dubited.706362

ABSTRACT

The levels of metals associated with dust is higher in indoor environment as compared to settled dust or soil in the exterior counterpart in the urban centers. The metals can be transferred to human body via inhalation, ingestion and dermal contact upon exposure and pose a significant health problem. The primary objectives of this study are (i) to determine the levels of metals in home dust samples in Bolu, Turkey, (ii) to assess the associated health risk when citizens are exposed to these metals in indoor environment. To end this, sixteen vacuum cleaning bags containing dust were collected from the homes located in the city center of Bolu (Turkey) between November and December 2017. The collected samples were analyzed by employing Wavelength Dispersive X-Ray Fluorescence (WDXRF) spectrometer in terms of major (Al, Ca, Cl, K, Mg, Na, P, S and Si) and minor (As, Ba, Br, Ce, Co, Cr, Cu, Fe, Mn, Nb, Ni, Pb, Rb, Sn, Sr, Ti, Y, Zn and Zr) metals at Turkish Atomic Energy Agency, Radiation and Accelerator Technologies Department, Ankara (Turkey). The measured levels of metals in the samples were ranged from 6.52±1.60 $\mu g\ g^{-1}$ for Y to 10.4±3.3 % for Na. The crustal enrichment factor (EF_{crust}) was calculated in order to understand the contamination level of household dust samples as compared to soil composition. EF_{crust} results revealed that there is minimal enrichment of Si, Rb, Ti, Ba, K, Y and Mn in household dust samples with respect to soil composition. On the other hand, Zn, Cl, and S found to be extremely enriched in the samples according to EF_{crust} values. Health risk assessment due to household dust metal exposure depicted that ingestion of dust particles is the main route of exposure for both adults and children. Overall, the calculated HQ value <1.0 suggesting there is no significant non-carcinogenic health risk for the residents. Cancer risks associated with Pb and Cr were estimated to be within the EPA's safe limits ($1x10^{-6}$ and $1.0x10^{-4}$).

Keywords: *Household Dust, Metals, WDXRF, Health Risk, Bolu*

Bolu'da Ev Tozu ile İlişkili Sağlık Risklerinin Tahmin Edilmesi

ÖZET

Kent merkezlerinde iç ortamda bulunan tozun metal içeriğinin dış ortamda çökelmiş halde bulunan toz ya da toprak içeriğine kıyasla daha yüksek olduğu bilinmektedir. Metaller insan vücuduna solunum, yutma ve deri teması ile alınabilmekte ve çok ciddi sağlık sorunlarına neden olmaktadır. Bu çalışmanın temel amaçlarını (i) Bolu'da evlerden toplanan tozun metal içeriğinin belirlenmesi, (ii) iç ortamda bu metallere maruz kalan bireylerin sağlık riskinin hesaplanması, olarak özetleyebiliriz. Bu amaçla, Kasım-Aralık 2017 tarihleri arasında Bolu şehir merkezinde bulunan on altı farklı evden toz içeren süpürge torbaları toplanmıştır. Toplanan örnekler Türkiye Atom Enerjisi Kurumu, Radyasyon ve Hızlandırıcı Teknolojileri Departmanı'nda (Ankara) Dalgaboyu Kırınımlı X-

Received: 19/03/2020, Revised: 01/09/2020, Accepted: 03/09/2020

Işınları Floresan Spektrometre (WDXRF) cihazı ile majör (Al, Ca, Cl, K, Mg, Na, P, S ve Si) ve eser (As, Ba, Br, Ce, Co, Cr, Cu, Fe, Mn, Nb, Ni, Pb, Rb, Sn, Sr, Ti, Y, Zn ve Zr) metalleri açısından analiz edilmiştir. Ölçülen metal seviyelerinin 6.52±1.60 µg g^{-1} (Y) ile %10.4±3.3 (Na) arasında değiştiği belirlenmiştir. Toz örneklerinin toprağa göre zenginleştirme faktörü (EFcrust) hesaplanarak örneklerin metaller açısından kontaminasyon düzeyi de bu çalışmada incelenmiştir. Elde edilen sonuçlar Si, Rb, Ti, Ba, K, Y ve Mn metallerinin toprak kompozisyonuna yakın seviyelerde olduğu belirlenirken, Zn, S ve Cl metallerinin toz örneklerinde belirlenen seviyelerinin toprak komposizyonuna kıyasla oldukça yüksek olduğu saptanmıştır. Toz örneklerinin metal seviyeleri dikkate alınarak yapılan sağlık riski değerlendirmesi hem çocuklar hem de yetişkinler için yutmanın ana maruziyet yolu olduğunu göstermiştir. Hesaplan HQ değerinin birden küçük olması, tozda bulunan metallerin bireylerin kanser olmayan risklere maruziyetinin önemli olmadığını işaret etmektedir. Kurşun ve Cr seviyeleri dikkate alınarak hesaplanan kanser riskinin ise EPA'nın güvenilir limiti aralığında ($1x10^{-6}$ ve $1.0x10^{-4}$) olduğu belirlenmiştir.

Anahtar Kelimeler: *Ev Tozu, Metaller, WDXRF, Sağlık Riski, Bolu*

I. INTRODUCTION

Human exposure to indoor contaminants has been attracting the attention of scientists since people spend up to 90 % of their time indoors [1]. Household dust is a heterogeneous mixture of materials from various sources, which consists of tracked-in or re-suspended soil particles, clothing, atmospheric deposition of particulates, hair, natural or artificial fibers, molds, pollen, allergens, bacteria, viruses, ash, soot, animal fur and danger, smoke, skin particles, cooking and heating residues, building constituents and among others [2].

Household dust can be a significant exposure route for some substances. On the other hand, majority of these substances do not pose a significant health problem as long as their corresponding concentrations are below the threshold levels. Among the metals, lead (Pb), arsenic (As) and cadmium (Cd) are known to have potential health risk once their concentrations exceed the threshold values particularly for the young children [3]. They can ingest appreciable quantity of household dust via hand to mouth and object to mouth activities. The ingestion of household dust by children is significantly high proportionate to their lower body weight. Moreover, human exposed to the contaminants associated with the household dust via inhalation and dermal contact. On the other hand, the previous studies have suggested that oral ingestion is the main exposure routes for humans as compared to dermal contact and inhalation. Ingested dust accesses to the gastrointestinal track, where metals partially dissolve and enter to circulatory system and carried to the tissues and organs of the human body [4].

Metals present in trace levels in many environmental compartments including natural water, air, dusts, soils, and sediments, which play a crucial role in human life [5][6]. Atmospheric fall out of petrol, tire wear, corrosion of metallic parts of the automobiles, roof tiles, paint and release from carpets and smoking can be considered as the major sources of trace elements in household dust [7]. More recently, cooking has been also shown as an indoor particle source [8][9][10][11]. The long-term exposure to metal contaminated household dust can affect the human health due to their toxicity, persistence and accumulation characteristics. Cadmium, Cr, Cu, Pb and Mn are considered as substances in household dust with potential risk to adults while Al, As, Cd, Cr, Cu, Pb, Mn, Ni and Zn are the metals in home dust with potential risk to children [3]. Lead has a half-life of 4 years in the human body and can stay up to 10 years in the bones upon exposure. Needleman (2009) [12] has put forward that Pb is destructive for the nervous system, kidney, circulatory and reproductive systems, particularly for children. The half-life of Cd in the human body is 6.2 to 18 years and this metal is known as its neurotoxic effect and adverse impacts on kidney [13]. Zinc, Cu, Mn, Cd and Pb are reported as essential human nutrients for human. However, these two metals can be initiators or promoters of carcinogenic activities in animals [14]. Adequate intake of Cu protects against Pb, on the other hand, excess intake of this metal leads to Pb absorption in human body [15][16]. Its higher intake results in its involvement in the generation of highly reactive species, for example, hydroxyl radicals, which are known as their devastating effects in cells, particularly, DNA damage and oxidation of proteins and lipids [17]. Long-term or higher dosage treatment of Zn has been shown to deplete the Cu in the human body [18][19]. Duda-Chodak and Blaszczyk (2008) [20] reported that impacts of Ni exposure and intoxication result in the dermatitis, skin allergies, pulmonary fibrosis, and cardiovascular and kidney disease. Chromium presents in nature in different oxidation states while the most commons are Cr (III) and Cr (VI). Exposure to Cr leads to dermatitis, allergies, as well as respiratory, gastrointestinal, neurologic, and reproductive problems, and cancer. Stahl et al. (2017) [21] claimed that Al influences the hematopoietic and nervous systems and the skeleton, triggering hemodialysis encephalopathy, anemia, aluminosis, osteomalacia, and osteoporosis, among other adverse health problems. Iron is known as the second most plentiful metal in environment and is crucial to a number of biological processes. The accumulation of Fe in the body tissues results in the cirrhosis, liver carcinoma, heart failure, diabetes mellitus, and osteoporosis [22].

The health risk posed by the contaminants in household dust on the residences has been studied in different cities over the world [23] - [27]. There are few studies in Turkey that investigated the human health risk assessment of chemical substances associated with the household dust. Kurt-Karakus (2012) [23] collected bags of the vacuum cleaners from 39 homes in mega-city Istanbul (Turkey). The levels of Cu, Pb, Cd, Zn, Cr, Mn, Co and Ni were determined in these samples. Moreover, Dündar et al. (2011) [28] reported deposition rates of heavy metals in the indoor dust, however, the concentration values were not provided in these studies. To our knowledge, this study is the first in Turkey that reported the levels of large suite of metals (Al, Ca, Cl, K, Mg, Na, P, S, Si, As, Ba, Br, Ce, Co, Cr, Cu, Fe, Mn, Nb, Ni, Pb, Rb, Sn, Sr, Ti, Y, Zn and Zr) in household samples. The objectives of this study are (1) to determine the levels of metals in household dust samples in Bolu, Turkey, and (2) to assess the associated health risk when citizens are exposed to these metals in indoor environment.

II. MATERIALS & METHODS

A. 1. Sampling

Bags of the vacuum cleaners were collected from the 16 houses located in the Bolu city center between November and December 2017. Though Harrad et al. (2006, 2008) [29], [30] followed a specific protocol for vacuum sampling of household dust from a certain area for a known time duration, vacuum cleaning bags used in the houses were received in this study in order to collect more representative samples for the houses and to be able to compare with the previous studies. Therefore, the level of metals given in this study will provide a rough idea about the pollution profile of the houses. The collected bags were put into the nylon bags, labeled and transferred to the laboratory. The dust from the bags were screened to remove any visible hair, soil, and grit from the samples manually by using the plastics tweezers and then sieved through a 100 mesh polystrene sieve, dried at 20 ºC and kept in the nylon bags till analysis. Figure 1 depicts the locations of the sampling sites. Moreover, the homeowners were asked some questions regarding the characteristics of their homes and these questions and given answers were provided in Table 1 below.

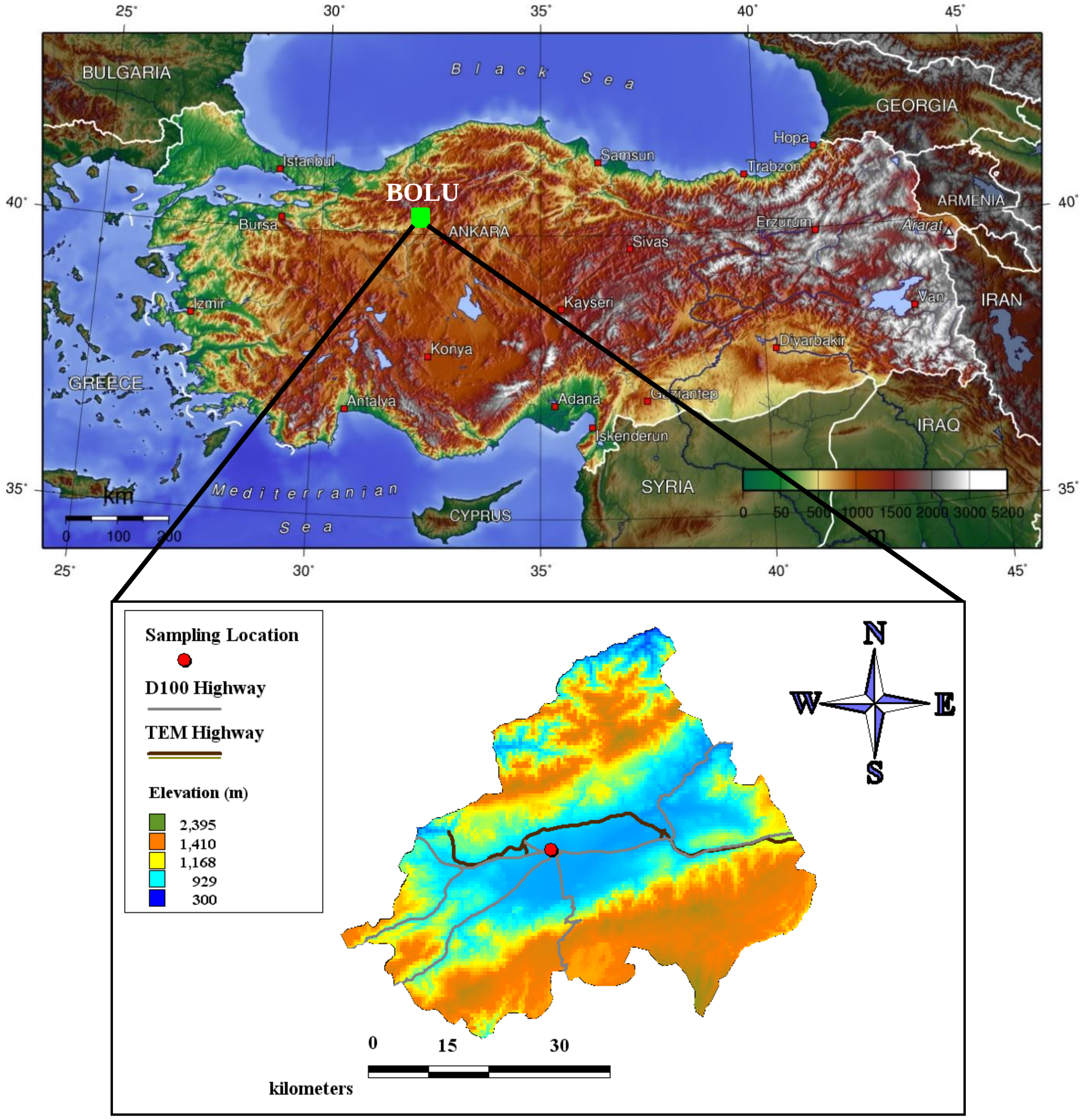

Figure 1. *Sampling area (elevation map of Turkey was taken from wikipedia [31], Bolu city map was taken from [32])*

Table 1. *The characteristics of the homes where dust samples collected*

Questions	Answers (Number of samples in each category)					
What is the characteristics of the sampling area?	Urban	Suburban	Rural			
What is the age of the building?	< 5 years (4)	6-10 years (4)	>10 years (3)	No Idea (3)		
What is the size of the home?	< 100 m^2 (4)	100-150 m^2 (4)	>150 m^2 (2)	No Idea (4)		
What is the structure of the building?	Concrete (14)	Wood	Shanty	Sun-dried brick	Steel	Others
What kind of fuel do you use for heating?	Natural gas (12)	Coal	Coal and Wood	Wood	Bottled gas	Others (2)
What kind of fuel do you use for cooking in the oven?	Natural gas (13)	Coal	Coal and Wood	Wood	Bottled gas (1)	Others
What is the floor cover of the house?	Polished wood (2)	Hardwood (1)	Laminate (11)	Tile	Vinyl	
What type of carpet do you use in the home?	Hand-knotted carpet (1)	Machine-made carpet (13)	Wall to wall carpet	Others		
Is there any people smoking in the house?	Yes (6)	No (8)				
What is the floor level of your home?	Ground floor (2)	1 (2)	2 (1)	3 (5)	4 (2)	5 (2)
Is your house close to main street?	Yes (11)	No (3)				
Is your house close to any kind of farm?	Yes (2)	No (12)				
Does your vacuum cleaner have bag?	Yes (9)	No (5)				
Do you have air conditioner in your home?	Yes (1)	No (13)				
Do you keep windows open during the day?	Yes	No (14)				
Has the house been repaired/painted in the last one year?	Yes (1)	No (12)				
Do you have pets in the home?	Yes	No (14)				

A. 2. WDXRF Analysis

The samples were dried at 105 ºC for 2 hours and 3-5 g of samples were weighed. The 3636 X-Press®, which is a 30-ton hydraulic laboratory pellet press, was used to make sample pellets for XRF analysis. Then samples were analyzed by using Panalytical Axios Adwance model WDXRF spectrometer in Radiation and Accelerator Technologies Department of Saraykoy Nuclear Reseach and Training Center of Turkish Atomic Energy Agency (Ankara, Turkey). Various environmental samples, including ambient particulate matter (PM) filters [32], have been analysed with this instrument and hence, procedure followed is well established. An X-ray tube (Imax= 160 mA, Vmax= 60 kV) with a power of 4.0 kW and Rh anode (SST-mAX) was employed in the WDXRF spectrometer. The samples were analysed for Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Nb, Ni, P, Pb, Rb, S, Si, Sn, Sr, Ti, Y, Zn and Zr. L-α line was used for Ba and Pb, and K-α line was used for the rest of the parameters. SUPERq OMNION software was used during the analysis of major elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti and Fe) while trace elements were analysed with ProTrace software. The duration of major and trace element analyses was 20 and 60 minutes, respectively. Proportional counter detector was used for the analyses of elements up to Ce in the presence of P10 gas mixture. On the other hand, NaI detector was used for the elements > Cr. Only 27 mm diameter sections of the dust samples were exposed to X-rays, and counting results were area-corrected. The standard reference materials (SRMs), currently available in the market, do not have a large suite of elements, for this reason, a combination of SRMs was used in this study to check the accuracy of the measurements. The accuracy of the WDXRF analysis was checked by analysing SRM 2703-Sediment for Solid Sampling, SRM 8704-Buffalo River Sediment and SRM 1646a-Eastaurine Sediment (NIST, USA). In addition, PTXRFIAEA09 sample prepared by the International Atomic Energy Agency (IAEA) and used for the inter-laboratory comparison of 45 laboratories for XRF measurements was employed in this study to find the accuracy. Method detection limit (MDL) values were automatically calculated by the spectrometer upon the analysis of SRMs. In addition, precision of the measurements was calculated as percent relative standard deviation (RSD) using repeated analyses of the SRMs. QA/QC results corresponding to XRF measurements were tabulated in Table 2. The uncertainty of the measurements were calculated based on the uncertainties associated with the (i) calibration curve, (ii), sample preparation, (iii) statistical count errors, (iv) stability of spectrometer, and (v) SRMs used for the accuracy calculation. The expanded uncertainty (k=2) values were provided in Table 2.

A. 3. Crustal Enrichment Factor (EF_{crust}) Calculation

The crustal enrichment factor (EF_{crust}) was calculated for the parameters measured in the home dust samples in order to understand the contamination rate of home dust samples with respect to natural soil composition. In this method, the ratio of concentrations of elements in home dust samples to a reference element is compared to the same ratios of geological materials as given in Equation 1:

$$EF_{crust} = \frac{(\frac{C_x}{C_{Al}})_{sample}}{(\frac{C_x}{C_{Al}})_{reference}} \quad (1)$$

The Mason soil composition was used in Equation 1 and "Al" was used as the reference element since it is the most common one in the Mason soil composition [33]. The categorization defined in Sutherland (2000) [34] was followed in this study. The EF_{crust} value <2.0 indicates that minimal enrichment in terms of metals. EF_{crust} value between 2.0 and 5.0 represent that dust samples were moderately enriched. Calculated EF_{crust} values from 5.0 to 20 and from 20 to 40 indicates significant and very high enrichment of dust samples, respectively. EF_{crust} >40 implies that dust samples extremely enriched.

Table 2. *QA/QC parameters in WDXRF analysis*

Parameter	**LOD**	**Precision**	**Accuracy**	**Uncertainty (k=2)**
	(*ppm*)	***(%)***	***(%)***	***(%)***
Al	134	0.38	2.04	3.75
Ca	19	1.36	1.19	4.46
Cl	25	3.21	3.76	9.12
Fe	55	0.42	2.00	3.74
K	46	1.38	3.80	4.29
Mg	78	1.20	4.56	3.93
Mn	12.66	0.73	5.47	4.43
Na	128	2.44	4.82	5.91
P	20	2.77	3.13	7.89
S	35	2.43	2.89	8.95
Si	350	0.48	1.70	4.40
Ti	19	0.63	1.40	7.62
As	4.88	3.13	2.83	8.81
Ba	5.68	1.98		5.98
Br	0.48	1.56		2.79
Ce	18.1	2.10		7.12
Co	3.51	3.03	1.57	4.00
Cr	1.50	2.13	9.59	1.92
Cu	0.80	3.73	4.43	5.85
Ga	0.61	1.61		2.05
La	7.04	3.85	6.97	11.94
Nb	0.50	0.60	7.70	5.27
Nd	11.6	3.68		8.42
Ni	0.80	1.72	4.04	2.24
Pb	1.38	0.82	5.55	4.70
Rb	0.40	0.46	3.35	0.95
Sc	1.43	1.78	0.48	8.73
Sr	0.30	0.64	2.05	0.91
Th	1.60	1.48	16.04	12.79
U	1.32	2.74	3.73	10.94
V	2.40	2.76	6.27	2.43
Y	0.60	0.62	4.09	1.77
Zn	0.60	0.74	4.96	1.40
Zr	0.50	0.67	7.99	0.87

A. 4. Health Risk Estimation

The model, which was developed by US Environmental Protection Agency [35], was used in this study to calculate the exposure of children and adults to metals determined in household dust samples collected in Bolu. Children and adults are exposed to dust through three main pathways: (1) ingestion of dust particles, (2) inhalation of dust particles via nose and mouth and (3) dermal contact [36][37]. The chemical daily intake (CDI) through each of the pathways was calculated by deploying the Equation (2), (3) and (4) as given below [38], [35], [23]. The parameters used in Equation (2), (3) and (4) and their corresponding values are provided in Table 3.

$$CDI_{ing} = C_{UCL} \times \frac{R_{ing} \times F_{exp} \times T_{exp}}{ABW \times T_{avrg}} \times 10^{-6} \qquad (2)$$

$$CDI_{inh} = C_{UCL} \times \frac{R_{inh} \times F_{exp} \times T_{exp}}{PEF \times ABW \times T_{avrg}} \qquad (3)$$

$$CDI_{dermal} = C_{UCL} \times \frac{SAF \times A_{skin} \times DAF \times F_{exp} \times T_{exp}}{ABW \times T_{avrg}} \times 10^{-6} \qquad (4)$$

"C_{UCL}" in above equations stands for the exposure-point upper confident limit content in terms of µg g^{-1} and corresponds to the upper limit of 95 % confidence interval of the mean concentration value, which is assumed to produce an estimate of the "reasonable maximum exposure" [39], [37], [40]. "Adjusted Central Limit Theorem" was utilized to calculate the 95 % of the upper confidence limit (UCL) of the metal concentrations in dust samples (Equation 5) as associated data conform to non-normal distribution in the present study.

$$C_{UCL} = \bar{X} + \left[Z_\alpha + \frac{\beta}{6\sqrt{n}}(1 + 2 * Z_\alpha^2)\right] * \frac{STD}{\sqrt{n}} \qquad (5)$$

In Equation (5), $\bar{X}$ is mean value of the data, n is the number of samples, STD stands for the standard deviation of the data, ß is the skewness of the data and $Z\alpha$ is the $(1-\alpha)^{th}$ quantile of the standard normal distribution, which equals to 1.645 at 95 % confidence level.

The hazard quotient (HQ) is calculated by multiplying the calculated CDI values corresponding to each exposure pathway for each element with BAF, the ratio of concentration of bioavailable metal to total metal concentration in dust samples, and then divided by the RfD_0, an estimation of the maximum permissible risks to human population through daily exposure by taking sensitive groups during their lifetime into consideration (Equation 6) [23]. The HQ value between 10^{-4} and 10^{-6} implies acceptable non-carcinogenic risks [38]. The hazard index (HI) is the sum of all HQ values for a particular metal (Equation 7). If the calculated HI > 1.0, there are potential non-cancer risks upon metal exposure. Otherwise, experiencing chronic risks is assumed unlikely. Carcinogenic risk associated with the metals in dust samples is calculated by multiplying the BAF with the slope factor (SLF) as given in Equation 8 [23].

$$Hazard\ Quotient (HQ) = (CDI \times BAF)/RfD_0 \qquad (6)$$

$$Hazard\ Index\ (HI) = \sum HQ_i \qquad (7)$$

$$Carcinogenic\ Risk = CDI_{ing,inh,dermal} \times BAF \times SLF \qquad (8)$$

Table 3. *Parameters used for the estimation of human health risk assessment from household dust*

Parameters	**Description**	**Unit**	**Value**		**Reference**
			Adult	**Children**	
$\mathbf{C_{UCL}}$	Metal Concentration in dust	$\mu g\ g^{-1}$			This Study
$\mathbf{R_{inh}}$	Inhalation rate	$m^3 day^{-1}$	20	7.6	[41], [37]
$\mathbf{R_{ing}}$	Ingestion rate	$mg\ day^{-1}$	100	200	[42], [37]
$\mathbf{F_{exp}}$	Exposure frequency	$day\ year^{-1}$	180	180	[36]
$\mathbf{T_{exp}}$	Exposure duration	years	24	6	[42], [37]
$\mathbf{A_{skin}}$	Skin area	cm^2	5700	2800	[42], [37]
SAF	Skin adherence factor	$mg\ cm^{-2}h^{-1}$	0.7	0.2	[42], [37]
DAF	Dermal absorption factor	-	0.001	0.001	[37]
PEF	Particle Emission Factor	$m^3 kg^{-1}$	1.36E+09	1.36E+09	[42]
ABW	Average Body Weight	kg	70	15	[42], [37]
$\mathbf{T_{avrg}}$	Averaging time	day			
	Carcinogens		25550	25550	[36], [37]
	Non-carcinogens		T_{exp}x365	T_{exp}x365	[36], [37]

III. RESULTS & DISCUSSION

A.1. General Characteristics of Data

Household dust samples collected in Bolu city center were analyzed in terms of metals (Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cu, Fe, K,m Mg, Mn, Na, Nb, Ni, P, Pb, Rb, S, Si, Sn, Sr, Ti, Y, Zn and Zr) in the current study. The descriptive statistics of the measured parameters were provided in Table 4. The measured levels were ranged from 6.52±1.60 µg g^{-1} for Y to 10.4±3.3 % for Na. Among the metals analyzed, As was detected in only one sample with a concentration value of 27 µg g^{-1}.

Table 4. *Summary statistics of measured parameters*

	Unit	N	Avg	SD	Median	Geomean	Min	Max
P	%	16	0.187	0.093	0.160	0.169	0.075	0.463
K	%	16	1.03	0.29	0.95	1.00	0.60	1.55
Cl	%	16	1.22	0.59	1.29	1.05	0.22	2.33
S	%	16	1.32	0.46	1.32	1.25	0.61	2.35
Al	%	16	1.37	0.62	1.18	1.24	0.59	2.70
Mg	%	16	3.88	1.16	3.66	3.72	2.08	6.67
Si	%	16	4.05	1.67	3.54	3.76	2.04	7.78
Ca	%	16	7.34	1.49	7.09	7.18	3.76	9.83
Na	%	16	10.4	3.3	10.2	9.8	4.6	16.7
Y	µg g^{-1}	8	6.52	1.60	6.59	6.33	4.16	9.03
Nb	µg g^{-1}	1	8.17		8.17	8.17	8.17	8.17
Rb	µg g^{-1}	15	14	3	14	14	9	20
Br	µg g^{-1}	15	20	29	11	13	5	120
As	µg g^{-1}	1	27		27	27	27	27
Co	µg g^{-1}	5	30.4	7.8	30.2	29.5	19.5	40.9
Ce	µg g^{-1}	2	64	41	64	57	35	93
Zr	µg g^{-1}	11	83	48	77	72	26	192
Ni	µg g^{-1}	16	89	130	63	61	24	571
Pb	µg g^{-1}	11	107	240	41	41	11	830
Sn	µg g^{-1}	3	123	138	65	75	23	280
Sr	µg g^{-1}	16	168	42	170	163	100	285
Ba	µg g^{-1}	13	183	256	104	119	44	1010
Cr	µg g^{-1}	16	197	138	152	158	49	502
Mn	µg g^{-1}	16	254	52	241	249	158	361
Cu	µg g^{-1}	15	271	577	94	119	23	2330
Zn	µg g^{-1}	16	752	761	551	523	92	3070
Ti	µg g^{-1}	16	827	367	850	700	112	1640
Fe	µg g^{-1}	2	8845	3189	8845	8553	6590	11100

The levels of metals found in the dust samples collected in the current study were compared with the values reported in the literature (Table 5). Among the measured metals, the highest values were determined for Na, Mg, S, Ca, Cr, Co and As in this study as compared to other studies used in the comparison. Lanzerstorfer (2017) [43] has also determined the relatively higher concentrations of Ca, Mg, Co and S in the household dust samples in his study. In addition, author identified increasing concentration of Co and S with decreasing size fractions, which implies that these metals are associated with the anthropogenic origin. It is worth the mention here that Khan et al. (2019) [44] reported As level in an industrial area in Bangladesh as 8.9 ppm, which is almost three times smaller than corresponding value measured in this study. Arsenic was detected in only one sample in this study and the dust sample was collected from a house in which coal and biomass are used for domestic heating. Consequently, source of As in this dust sample can be attributed to the coal combustion. Moreover, Rasmussen et al. (2018) [45] reported that paint pigments contain Cr and wood used in the building materials is also treated with As and Cr. *Kurt-Karakuş (2012)* [23] collected dust samples from houses located in urban, suburban and rural locations in Istanbul and median concentration were given in Table 5 for comparison. Among the common metals used in the comparison, only Zn and Ni levels reported by Kurt- Karakuş (2012) [23] are higher than ones determined in Bolu. Reis et al. (2019) [46] reported that environmental sources of Zn are vehicular emissions, biomass burning and industrial activities. Moreover, the smoke related sources for instance, cooking, wood burning in stoves and fireplaces, agricultural waste burning and forest fires are reported to be the sources of Ni in the same study. Cempel and Nikel (2006) [47] suggested that combustion of coal, diesel oil and fuel oil, the incineration of waste and sewage contributes to the ambient Ni levels while stainless steel utensils in the kitchen, tobacco smoking and inexpensive jewelry are accounted among the indoor Ni sources. Al-Rajhi et al. (1996) [48] revealed that 93 % of the Ni and Zn among other metals in indoor dust is originated from outdoor dust and 4% of this is due to automobile emissions. Consequently, the relatively higher levels of Ni and Zn in Istanbul could be attributed to higher anthropogenic activities in this mega-city of Turkey. The lowest Rb concentration was detected in this current study as compared to other studies [49] [44]. Figueiredo et al. (2007) [50] reported that the main source of Rb in the ambient air is soil. Therefore, Rb may probably be transported to indoor with human activities and ventilation. Other metals (Al, K, Ti, Mn, Fe, Cu, Pb, Ba and Sr) measured in the current study have comparable concentrations with the values reported in other studies given in Table 5.

Table 5. *Comparison with the relevant literature (Unit: µg g^{-1})*

References	This Study	[23]	[51]	[52]	[53]	[54]	[43]	[49]	[44]	[55]
Place	**Urban (Bolu, Turkey)**	**Urban (Istanbul, Turkey)**	**Urban (Al-Karak, Jordan)**	**NS* (Sydney, Australia)**	**Industrial (Ahvaz, Iran)**	**Urban (Shanghai, China)**	**Urban (Wels, Austria)**	**NS (Ottawa, Canada)**	**Industrial (Bangladesh)**	**Urban (Cairo, Egypt)**
Na	103719						7610	23224		
Mg	38781						7050	9826		
Al	13678					1650	3703	25948		1524.9
P	1873									
S	13224						10770			
K	10337						2460	10305	10892	
Ca	73369						66700	48760	30898	
Ti	827		291.3				102		4349	
Cr	197	55	72.5	90	15 (26)	16	31	86.7		77.63
Mn	254	136	243.2	220	93 (139)	125	234	269		
Fe	8845					1620	5167	14135	57435	2691.5
Ni	89	263	70	50.9	11 (20)	41.1	49	62.9		77.69
Cu	271	156	90.4	272	115 (159)	175	190	206	24.9	124.76
Zn	752	832		1876	696 (890)	695	395	716.9	1717.9	190
Pb	107	28	51.9	299	63 (84)	63.1	29	405.6	310.5	321.96
Ba	183					276	66	492		
Br	19.6									
Rb	14.1								126.3	
Sr	168						57	255	154.5	
Co	30.4	5			9.5 (11.5)	1.78				3.83
As	26.5		2.6	17.6		3.5			8.9	
Sn	123									

A.2. Metal Sources

Pearson correlation coefficient analysis of the measured parameters in household dust samples were performed by using Excel in Microsoft Office Professional Plus 2016 in this study. The results of the correlation coefficient analysis showed that there was a significant and positive correlation among Al, Br, Mn and Si ($p<0.005$). Aluminum and Mn in household dust were previously ascribed to track-in soil particles from outdoor by Yoshinaga et al. (2014) [25]. Calcium is known as one of the major component of natural soils but it showed only statistically significant correlation with Sr (R> 0.72, $p <0.005$). Larsen et al. (2015) [56] have revealed that Ca has been used in many consumer products ranging from food to cosmetics. In addition, wall dust (that is, coatings and building materials) can be a source of Ca in indoor PM [57]. Khan et al. (2019) [44] also found statistically significant positive correlation between Ca and Sr in household dust, indicating the same origin of these metals. Though S is known as marker of coal combustion, it also depicted positive statistically significant correlation with Sr (R> 0.79, $p <0.001$). Calderon et al. (2017) [58] found that consumer spray products releases Sr as well as other metals to indoor air, which contributes to $PM_{2.5}$ mass. According to the literature reviwed, it can be concluded that Ca, Sr and S could be originated from the consumer products used in the homes. Chlorine, Na and K had significantly positive correlation (R=0.88, $p<0.001$) while Cl and K showed negative moderately strong correlation with Si (R=-0.60, $p <0.01$). On the other hand, Al and Si depicted significant positive correlation (R=0.86, $p <0.01$). Chlorine, Na, K, Al and Si are major component of earth crust [59]. The positive strong correlation of Si with Al and its negative correlation with Na, K and Cl revealed that Si has same source with Al but not with the rest of the earth crust metals determined in this study. Conner et al. (2001) [60] put forward that cosmetics and personal hygiene products are the sources of Al and Si to indoor dust in addition to many others including Bi, Ti, Mg and Fe. Moreover, it has been reported by Ciacci et al. (2015) [61] that aluminum silicates are common in kitty letters and food additives; Al is used in packaging and building/construction materials. In addition, pharmaceuticals, cosmetics, catalysts, pigments and paints include other aluminum compounds such as oxides, chlorides and sulfates. Consequently, it can be concluded that Al and Si have anthropogenic and common source in this study. On the other hand, the source of Na and Cl could be attributed to the sea salt used during cooking in the houses as suggested by Schauer et al. (1999) [62]. Barium is strongly correlated with Pb and Zn (R> 0.86, $p <0.001$). The source of Pb in indoor dust varies from Pb-based paint sources to accumulation of automobile related petrol Pb depositions [52]. Barrio-Parra et al. (2017) [63] revealed that the room in which the dust sample collected was a strong determinant of Zn levels. In addition, Zn in dust samples was shown to be highly correlated with the presence of smokers inside the home. Zinc has been used in the spare parts of the vehicles as Zn alloy and galvanized components. Hence, considerably amount of Zn is associated with the street dust due to abrasion of tires and other parts of the vehicles [64]. Accordingly, it can be concluded that the human facilities inside the home and dust adhere to foot are among the sources of Zn. Barium, Zn and Pb chromates have been used in paints as colored pigments [65]. Therefore, these metals have both outdoor and indoor sources affecting their levels in household samples. Among other anthropogenic elements, Cu and Ni had strong positive correlation (R> 0.98, $p <0.001$). Copper and Ni can be originated from local soil (natural soil) while their anthropogenic sources include corrosion of alloys used in the vehicle components, weathering of materials, such as paints and coatings, and other metal surfaces [64]. In addition, presence of Ni in household dust was previously associated with cigarette smoke, fuel consumption and chemicals used in the aerosol sprays [66].

The calculated EF_{crust} for the metals was depicted in Figure 2. Among the parameters analyzed, Si, Rb, Ti, Ba, K, Y and Mn have $EF_{crust} <2.0$ indicating minimal enrichment of dust samples with these metals. Low EF_{crust} values for the metals revealing that these metals may originate from soil or road dust resuspension. For Zr and Sr, EF_{crust} is between 2.0 and 5.0, hence, it can be concluded that dust samples were moderately enriched with these metals. Put it differently, these metals have both anthropogenic and natural origin. On the other hand, Ni, Cr, Pb, Mg, Ca and P have $5.0< EF_{crust} <20$, which implies that dust samples are significantly enriched with these metals. In other words, associated sources of these metals in household dust samples are not natural. Sodium, Br and Cu have $20< EF_{crust} <40$, which reveals very high enrichment of these metals in dust samples. Moreover, the calculated $EF_{crust} >40$ for Zn, S and Cl implying that these metals have anthropogenic origin in the dust samples. Hilger et al. (2013) [67] was able to identify chlorinated paraffin in house dust samples in Germany. Similarly, Shang et al. (2019)[68] measured the short and long chain of chlorinated paraffin in Canadian house dust. Li et al. (2019) [54] put forward that chlorinated flame retardants present in the household dust in Shanghai (China). Consequently, extremely high enrichment of Cl in dust samples collected in the current study may be associated with the organics in addition to inorganic dust and further detail analysis of samples for these parameters is emerging. Öztürk and Keleş (2016) [32] previously indicated that the source of S, Zn and Pb in the coarse fraction of PM in the ambient air of Bolu is a mixture of anthropogenic emissions and crustal material. Accordingly, one can conclude that Zn and S may have also outdoor source.

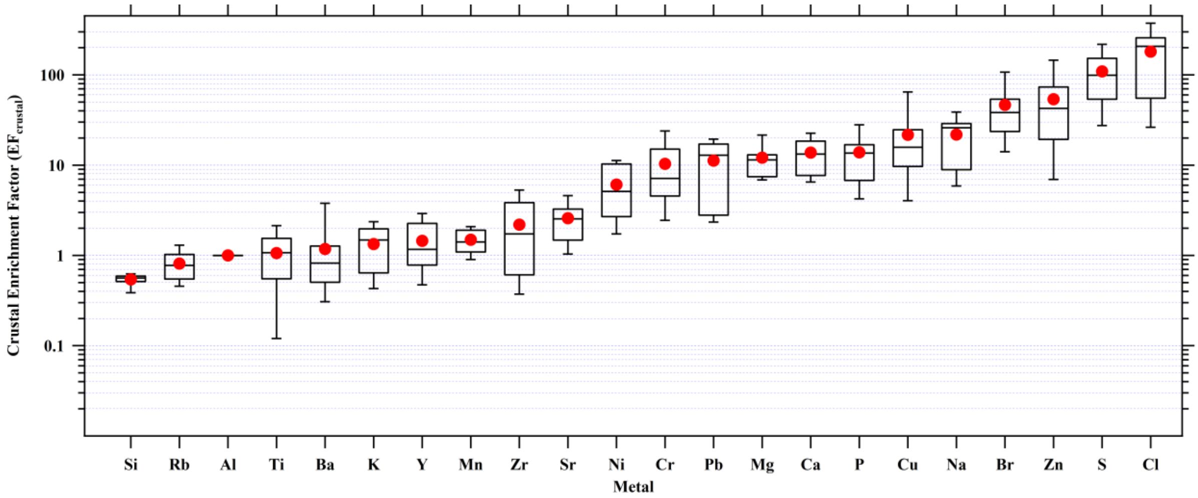

Figure 2. *Crustal EF_{crust} of the measured parameters*

A.3. Potential Health Risk of Heavy Metal Exposure

Carcinogenic and non-carcinogenic risks associated with the exposure of metals in household dust samples were calculated in this study and presented in Table 6. Ingestion was found to be the main route of exposure for children since calculated HQ_{ing}=1.56E-1, which is significantly higher than HQ_{dermal} and HQ_{inh}. The HQ_{ing} values for metals in indoor dust increase in the order of Ni<Zn<Mn<Cr non-canc. <Cu< Pb non-canc. as tabulated in Table 6. Similar to children, the main health risk associated with the metal exposure for adults is through ingestion. However, the calculated HQ_{ing} for adults is almost one fold smaller than that for children. In contrast to these, Öztürk and Keleş (2019*)*[69] found that dermal contact was the main route of exposure to metals associated with particulate matter in the ambient air at Bolu city center. The HQ_{ing} values for metals in indoor dust increase in the order of Zn<Mn<Ni<Cr non-canc. <Cu< Pb non-canc. as shown in Table 6 for adults. Overall, the HQ values are less than unity (HQ<1), revealing low non-carcinogenic risk to children and adults due to household dust metal exposure. The calculated EF_{crust} for Cu and Pb (> 20) suggests that the indoor dust is highly contaminated with these metals. Hence, the risks associated with these metals imposed on the children and adults is significantly higher than ones estimated for the other metals. The cancer risks associated with the Pb and Cr were also calculated in this study and corresponding results were provided in Table 6. The results revealed that cancer risks due to Cr and Pb exposed on the children is almost three times higher than ones for the adults, which can be attributed to the higher ingestion rate of metals among children. Ones the cancer risks for these metals compared, it can be concluded that the risks due to Pb is two times higher than Cr for both adults and children. Since calculated HI values are within the EPA's safe limits ($1x10^{-6}$ and $1x10^{-4}$), Cr and Pb do not impose cancer risk on adults and children living in Bolu city center. In order to perform more accurate health risk assessment of household dust associated metals posed on residents in Bolu, detailed chemical composition in terms of organics should also be evaluated in future studies.

Table 6. *Carcinogenic and non-carcinogenic risk associated with the metals determined in dust samples*

µg/g	Mn	Ni	Cu	Zn	Cr non-canc.	Cr canc.	Pb non-canc.	Pb canc.	Reference
C (95 % UCL)	255	92.12	273	754	199	199	110	110	This Study
R_fD_0 (mg/kg-day)	0.14	0.02	0.04	0.3	0.003		0.0035		[23]
BAF (%)	47.6	32.4	64.4	53.2	5.83		37.2		[23]
SLF (mg/kg-day)						0.50		0.28	[23] for Cr and [39] for Pb
Child									$HI = \sum HQ_i$
HQ_{ing}	5.71E-03	9.81E-03	2.89E-02	8.80E-03	2.54E-02		7.70E-02		1.56E-01
HQ_{dermal}	1.60E-05	2.75E-05	8.10E-05	2.46E-05	7.12E-05		2.16E-04		4.36E-04
HQ_{inh}	1.60E-07	2.74E-07	8.09E-07	2.46E-07	7.10E-07		2.15E-06		4.35E-06
$HI = \sum HQ_i$	5.73E-03	9.84E-03	2.90E-02	8.82E-03	2.55E-02		7.72E-02		1.56E-01
Cancer Risk						3.28E-06		6.48E-06	
Adult									$HI = \sum HQ_i$
HQ_{ing}	6.12E-04	1.05E-03	3.10E-03	9.42E-04	2.72E-03		8.25E-03		1.67E-02
HQ_{dermal}	2.44E-05	4.19E-05	1.24E-04	3.76E-05	1.09E-04		3.29E-04		6.65E-04
HQ_{inh}	9.00E-08	1.55E-07	4.56E-07	1.39E-07	4.01E-07		1.21E-06		2.45E-06
$HI = \sum HQ_i$	6.36E-04	1.09E-03	3.23E-03	9.80E-04	2.83E-03		8.58E-03		1.73E-02
Cancer Risk						1.46E-06		2.88E-06	

IV. CONCLUSIONS

The household dust samples were collected from Bolu city center in this study and analyzed with WDXRF for a large suite of metals. The measured levels were ranged from 6.52±1.60 µg g^{-1} for Y to 10.4±3.3 % for Na. Among the measured metals, the highest values were determined for Na, Mg, S, Ca, Cr, Co and As in this study as compared to other studies used in the comparison. Arsenic was detected in only one house and its concentration was 27 µg g^{-1}, which is three times higher than one reported for an industrial site in the literature. Zinc and Ni levels detected in this study were lower than ones measured in Istanbul. Considering the intensity of traffic in this mega-city, the higher concentration of these metals in indoor at Istanbul could be attributed to the traffic. The lowest Rb concentration was measured in household dust samples in the current study as compared to literature. The rest of the metals had comparable levels with the reported values in the literature. The low $EF_{crust}<2.0$ values for Si, Rb, Ti, Ba, K, Y and Mn suggest that these metals soil origin and transported to the indoor by adhesion on the foot. Except for the Zr and Sr, the rest of the metals have $EF_{crust}>5.0$, implying that there are several non-soil sources of these metals inside the homes. Lastly, the carcinogenic and non-carcinogenic risks associated with these metals were also evaluated in this study. The results revealed than ingestion of metals is the main route of exposure for both adults and children while the corresponding risk is almost one fold higher in the children as compared to adults. Copper and Pb are the two metals and their contribution to non-carcinogenic risks is higher relative to other metals. The calculated HQ < 1.0 for both adults and children implied that metals do not pose significant non-carcinogenic health risks on the residents of the homes at Bolu city center. Since calculated HI values are within the EPA's safe limits ($1x10^{-6}$ and $1x10^{-4}$), Cr and Pb do not impose cancer risk on adults and children living in the city.

ACKNOWLEDGEMENT: Authors thank to volunteer homes in this study for providing their vacuum cleaner bags for us. Moreover, authors are grateful to the undergraduate students of Environmental Engineering Department of BAİBU (Sahip Gök, Elif Kemal, Gizem Alagöz, Gülden Atalay, Tarık Arslan, İlyas Sezgin and Hamada Niyigena) for their helps in collecting the samples and filling the questionnaires at the visited homes, and preparing the samples prior to analyses.

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