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HEAVY METAL IN DIFFERENT SIZE FRACTIONS OF HOUSEHOLD DUST COLLECTED FROM RURAL RESIDENTIAL AREA OF SIMPANG RENGGAM, JOHOR

(Logam Berat dalam Pelbagai Pecahan Saiz Habuk Rumah dari Kawasan Kediaman Luar Bandar Simpang Renggam, Johor)

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Abstract

A study investigating the levels of selected heavy metals in household dust from a rural residential area was carried out. Household dust was collected from seven residential homes within the area of Simpang Renggam, Johor. All samples were sieved through a 200 µm sieve, in which two dust samples with sufficient masses (sample A and B) were further separated into four discrete fractions (<63 µm, 63-75 µm, 75-150 µm, and 150-200 µm) before analysis. Dust samples were acid digested and analyzed for the content of Al, Cr, Mn, Ni, Zn, Cu, Cd, Ba, Pb, Mg, and Fe using inductively coupled plasma - mass spectrometer (ICP-MS). Results showed that the Fe and Al were among the most abundant elements in bulk dust samples (mean concentration of 8500 mg/kg and 5100 mg/kg, respectively), and their concentrations increased with decreasing particle size. Mean concentrations of other elements ranged between 0.027 mg/kg to 2310 mg/kg. For Mn, Mg, Cu, and Zn, higher levels were measured in coarser particle size. Health risk estimation indicated that Hazard Quotient (HQ) and Hazard Index (HI) values were below than 1, representing no non-carcinogenic risk to the residents via ingestion, inhalation, and dermal absorption.

Keywords: indoor dust, heavy metals, particle size distribution, rural residential area

Abstrak

Satu kajian mengenai kepekatan logam berat terpilih dalam habuk dari kawasan perumahan luar bandar telah dilakukan. Sampel habuk dikumpulkan dari tujuh rumah kediaman di kawasan Simpang Renggam, Johor. Semua sampel diayak melalui ayakan 200 µm, di mana dua sampel debu dengan jisim yang mencukupi (sampel A dan B) dipisahkan menjadi empat pecahan diskrit (<63 µm, 63-75 µm, 75-150 µm dan 150-200 µm) sebelum analisis. Sampel habuk dicerna asid dan dianalisis untuk kandungan Al, Cr, Mn, Ni, Zn, Cu, Cd, Ba, Pb, Mg dan Fe dengan menggunakan teknik ICP-MS. Keputusan kajian menunjukkan bahawa Fe dan Al merupakan unsur yang paling banyak dalam sampel debu <200 µm (kepekatan purata sebanyak 8500 mg/kg dan 5100 mg/kg masing-masing), dan kepekatan unsur ini meningkat dengan penurunan saiz zarah. Purata kepekatan unsur-unsur lain berada dalam julat 0.027 mg/kg dan 2310 mg/kg. Untuk Mn, Mg, Cu, dan Zn, kepekatan yang lebih tinggi diukur dalam partikel habuk yang lebih kasar. Anggaran risiko kesihatan memberi nilai Darjah Bahaya (HQ) dan Indeks Bahaya (HI) kurang daripada 1, menunjukkan bahawa pendedahan penduduk kepada habuk melalui pengambilan, penyedutan dan penyerapan kulit tidak mendatangkan risiko (secara bukan karsinogenik).

Kata kunci: habuk rumah, logam berat, taburan saiz zarah, kawasan kediaman luar bandar

Introduction

People nowadays tend to spend most of their time indoors, especially during the COVID-19 pandemic period, giving rise to the concern regarding exposure to indoor contaminants. Dust in homes, offices, and floor may contain human and animal hair, paper fibres, and mineral from outdoor soil and many other materials which may be found in the local environment. The composition of indoor dust may vary greatly between rooms of a given house and among geographic locations [1]. House dust containing heavy metals could be generated indoors (e.g.: premises and furniture materials, consumer products, and resident activities) or tracked indoors from outside [2].

Studies investigating the contamination of heavy metals in dust as well as its potential health risk of exposure have been reported worldwide. Dust samples investigated in these studies were mostly of single particle size (such as <200 um) or without sieving. Generally, particle size of dust can vary, and the finest size can easily enter human body through inhalation, dermal absorption, and ingestion as it is less dense and electrostatically mobilized compared to the other big particle sizes. According to Lanzerstorfer [3], while the total surface area of the particles is indirectly proportional to the size of the particles, there is special interest in the size dependence of heavy metal concentrations. Since the transition of mass from a solid to a liquid takes place on the solid's surface, in cases with finer particles, the mass transfer rate is higher, thereby making hazardous components found in these finer particles more available. Therefore, a key consideration in the risk assessment of human exposure to heavy metals in indoor dust is particle size [4].

In Malaysia, a few studies have been carried out to determine the concentration of heavy metals in indoor dust from schools [5-7] and residential buildings [8], focusing on urban and semi-urban areas. However, data on metal composition in rural areas with low population density and main economic activities such as ecotourism and agriculture are still scarce. Therefore, the present study aimed to investigate the level of heavy metal in house dust collected from rural residential area of Simpang Renggam, Johor, according to dust particle size fractions.

Materials and Methods

Sample collection and treatment

The study area, Simpang Renggam (1.8273°N, 103.3100°E), is a small town located in the southern half of Kluang District, Johor, Malaysia. Seven vacuum cleaner dust samples were collected in August 2020 from different households of Simpang Renggam residential area. The dust samples were transferred into zip-lock bags, sealed, and labelled properly. The volunteers were required to answer a simple questionnaire regarding their household condition and personal behaviors (Table 1). Upon arrival at the laboratory, large objects such as hair, stone, and plastic object were discarded from dust sample using a tweezer before sieved through a 200 µm sieve. Due to a large sample mass required for particle size distribution analysis, only 2 out of 7 samples (Sample A and B) were suitable for further separation into four discrete fractions. The <200 µm portion was treated as the bulk dust sample, in which a portion of this <200 µm fraction dust was taken and analyzed for its total metal concentrations. The remaining portion was further fractionated using an Endecotts EFL 2000 sieve shaker into the following size fractions: <63 µm, 63-75 µm, 75-150 µm, and 150-200 µm, and each of these fractions were analyzed for metal concentrations. Due to limited amount of sample available for fraction 63-75 µm in Sample B (<0.1 g), the fraction was combined with the finest fraction, <63 µm, for chemical analysis.

Chemical and data analysis

All glass and plastic wares were cleaned with detergent, soaked in 5% (v/v) HNO_3 overnight and rinsed with deionized Milli-Q water before use. Approximately

0.2 g of dust sample were acid digested with 10 mL of H_2O_2: HNO_3 (3:7 v/v) using UltraWave™ system (Milestone Helping Chemist, Italy). The digestion program was set at 253 °C and 182 bar for 30 minutes, followed by cooling time with nitrogen gas for 10 minutes. After digestion, the sample solution was filtered with Whatman cellulose acetate filter paper and diluted to 50 mL with 2% HNO_3. For each batch of the digestion, one reagent blank was included. Concentration of aluminum (Al), chromium (Cr), manganese (Mn), nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), barium (Ba), lead (Pb), magnesium (Mg), and iron (Fe) were determined using inductively coupled plasma – mass spectrometer (Perkin-Elmer NexIon 300X). All statistical analyses (Spearman rank correlation, Mann Whitney *U* test, and Kruskal-Wallis *H* test) were performed using Excel 2016. The significance was set at $\alpha = 0.05$ in all the statistical analyses.

Health risk assessment

The average daily dose (ADD) of an element from dust via ingestion, inhalation, and dermal contact pathways can be estimated using Equations (1), (2) and (3), respectively, as suggested by the USEPA Exposure Factors Handbook [9]:

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad (2)$$

$$ADD_{der} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (3)$$

where ADD is exposure dose (mg/kg/day), C is the concentration of the element (mg/kg), IngR is the ingestion rate of contaminated dust for adult (30 mg/day), EF is the exposure frequency (350 day/year), ED is the exposure duration (30 year), BW is the body weight (70 kg), AT is the average time (ED×365 day), IngR is the inhalation rate (15.2 m^3/day), PEF is the inhalation factor for the respirable particles (1.36×10^9 m^3/kg), SA is the surface area of the skin exposed to pollutants (5700 cm^2), AF is the skin adherence factor (0.07 mg/cm^2/h), ABS is the dermal absorption factor (0.001).

Based on the ADDs from the three exposure routes, hazard quotient (HQ) and hazard indexes (HI) indicating the non-cancer risk during a lifetime can be calculated according to Equation (4) and (5):

$$HQ = \frac{ADD}{RfD} \quad (4)$$

$$HI = \sum HQ = HQ_{ing} + HQ_{inh} + HQ_{der} \quad (5)$$

where RfD is the estimated maximum permissible risk on humans through daily exposure (mg/kg/day). HQ and HI ≤ 1 indicates that adverse effects on human health are unlikely, while HQ and HI > 1 reveals probable adverse health effects.

Table 1. Description of the sampling sites

Household	**A**	**B**	**C**	**D**	**E**	**F**	**G**
Building type	terrace	terrace	Terrace	shop house	semi-detached	semi-detached	semi-detached
Size of the house (sqft)	~1200	~1200	~1200	~1200	<1000	~1200	~1200
Number of occupants	7	6	5	8	4	7	3
Age of the building (year)	15	37	8	25	25	15	28

Table 1 (cont'd). Description of the sampling sites

Household	A	B	C	D	E	F	G
Facing main traffic street	no	no	No	yes	no	yes	yes
Floor cover	tile	carpet	Tile	carpet	tile	carpet	carpet
Shoes allowed	no	no	No	no	yes	yes	no
Smoking allowed	no	no	Yes	no	yes	no	no
Air-conditioned	yes	yes	No	no	no	no	yes
Last painted (year ago)	<1	>5	1-5	1-5	1-5	1-5	1-5
Ventilation frequency	everyday	2-3 times a week	everyday	2-3 times a week	everyday	occasionally	occasionally
Pets allowed	yes	no	No	no	yes	no	no
Vacuuming frequency	once a week	once a week	once a week	once a week	once a week	once a week	once a week

Results and Discussion

Levels of heavy metals

The concentrations of heavy metals from indoor dust in Simpang Renggam, Johor are summarized in Figure 1. Among the eleven target metals, Fe showed the highest mean concentration (8500 mg/kg), followed by Al (5100 mg/kg) and Mg (2300 mg/kg). As the second and third most abundant element in the earth's crust, Fe and Al occur naturally in the environment, foodstuffs, and drinking water. Other possible sources of Fe and Al in indoor dust include unpaved roads and heavy construction activities of new buildings, as well as wind-blown dust from surface soil and road dust. Although the use of leaded petrol has been banned for many years in Malaysia, their residues are still detected in indoor dust samples, especially those collected from urban and sub-urban areas [5, 8, 10-11]. Cadmium was detected in one of the dust samples, probably derived from lubricating oils or old tyres [5] due to the absence of any significant industry near the study location. Compared to previous studies, the levels of heavy metals measured in this study were within the range of literature [12-16].

Spearman rank correlation tests were performed to identify correlations between individual elements in the samples. High positive correlations ($0.78 < r < 0.93$, $p < 0.05$) were found between Al-Ni, Al-Cu, Al-Ba, Cr-Cu, Cr-Fe, Ni-Ba, Ni-Pb, Zn-Mg, and Ba-Pb in dust samples (Table 2), suggesting a common source of origin for these metals. Similar correlations between Al-Ni and Cu-Cr have been reported by previous studies [10, 14]. On the other hand, concentrations of metals showed no relationship to house age (Spearman rank analysis, $p > 0.05$), whereas the concentration of Zn increase significantly with number of occupants (Spearman rank analysis, $r = 0.77$, $p < 0.05$). Associations between household conditions and personal behaviors for different metal concentrations in household dust were tested with Mann Whitney U test or Kruskal-Wallis H test as appropriate. However, the results revealed that neither household conditions (facing major roads, floor covered with carpet, and duration from the last painting) nor personal behaviors (ventilation frequency and houses that allow shoes, smoking, and pets indoors), were statistically significant for metal concentrations (Mann Whitney *U* test and Kruskal-Wallis *H* test, $p > 0.05$). This finding is contrary to previous studies which have suggested smoking [15, 17], natural ventilation frequency [17], and age of house [15, 18-19] as the important factors of heavy metals enrichment in household dust. This could be attributed to the small number of samples available for this study and the lack of other possible parameters in our questionnaire (e.g.: cooking frequency, type of fuel, paved or unpaved streets, number of windows, etc.).

Particle size distribution

Particle size distribution analysis was performed on two dust samples with sufficient sample mass. Both sample A and Sample B showed great similarity in the particle size distribution of dust (Figure 2). By considering the <200 μm size as the bulk sample (100%), both samples were dominated by the <63 μm fraction (39% and 42% of total dry mass, respectively), followed by 75-150 μm (35% to 37%), 150-200 μm (15% to 19%), and 63-75μm (2% to 11%).

Metal distribution with particle size

The concentrations of heavy metals in different particle size fractions are summarized in Table 3. In general, total metal concentrations decreased with particle size in Sample A, but sample B showed the contrary, giving higher total metal concentration in the coarser fraction. For both samples, the concentration of Al and Fe decreased with increasing particle size. This result is in agreement with several previous reports on the preferential partitioning of metals to fine particle size fractions [3, 20-22]. This observation could be explained by the greater surface area per unit mass of the fine particles, which increases the adsorption capacity of these fractions. For Mn, Mg, Cu, and Zn, on the other hand, higher concentrations were observed in the coarser fractions. The findings are contrary to Doyi et al. [23], who has reported the most concentrated metals in course size fractions (150-250 μm) for Cu, Ni, and Pb. However, Lanzerstorfer [3] found no distinct trend for the concentrations of Cu and Zn in different particle size fractions. For other remaining metals, no consistent trend could be observed regarding their distribution in different dust particle sizes, probably due to small sample size in this study, as well as the influence of geographical location and human activities in the chemical composition of household dust. Although different size fractions were analysed, the concentrations of Al, Cr, Cu, Ba, Pb, Mg, and Fe measured in the present study were within the range of values found in the literature [3, 15, 20-21]. Meanwhile, much higher levels of Mn and Zn were measured, whilst slightly lower concentrations of Ni were measured in this study. The proportions of metal loadings in different size fractions of dust samples are presented in Figure 3. The partitioning of most of the metals in different size fractions of Sample A was found to be consistent, as the finer fractions (<63 μm, 63-75 μm, and 75-150 μm) corresponded to >70% of the total metal concentration. For Sample B, it was evident that the metals were not homogenously distributed among the various particle's fractions. Cr and Ni were only detected in the finest fraction of Sample B (<75 μm), whereas more than 40% of the concentrations of Mn, Cu, Zn, and Mg resided in the coarse fraction (150 - 200 μm).

Exposure dose

The health effects caused by heavy metals exposure were assessed by using mean concentrations of bulk dust (<200 μm) according to Eq (1) – (5). The HQs and HIs calculated through three exposure pathways for adults are presented in Table 4. The HI values for non-carcinogenic effects decreased in the order of Zn>Al>Fe>Cr>Pb>Mn>Ba>Ni>Cu>Mg>Cd. In general, all of these HIs were less than 1, indicating that these concentration levels did not present a significant toxicity risk to the occupants. Exposure pathway via dust ingestion contributed to the highest risk, followed by dermal absorption, and inhalation. As shown in Table 3, metal concentrations in the smaller size fractions of dust may be 1-2 orders of magnitude higher than those measured in bulk dust, suggesting a possibility of lower estimates of inhalation exposure using <200 μm particle size.

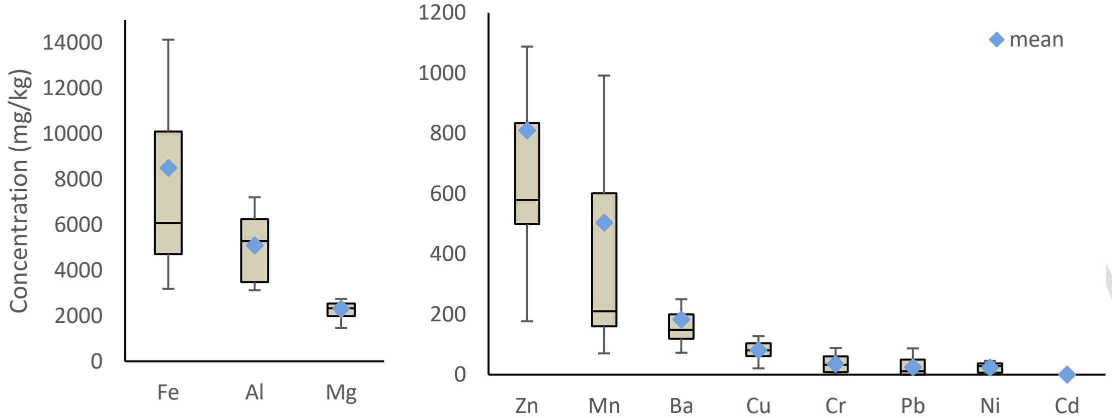

Figure 1. Concentration of heavy metals in vacuum cleaner dust samples

Table 2. Correlation matrix for heavy metal concentrations

	Al	**Cr**	**Mn**	**Ni**	**Zn**	**Cu**	**Cd**	**Ba**	**Pb**	**Mg**	**Fe**
Al	1	0.75	0.29	0.86*	0.14	0.82*	0.61	0.86*	0.74	-0.18	0.64
Cr		1	0.07	0.61	-0.11	0.93*	0.00	0.50	0.41	-0.39	0.79*
Mn			1	-0.07	0.71	0.32	0.41	-0.07	-0.11	0.68	-0.43
Ni				1	-0.14	0.57	0.61	0.93*	0.78*	-0.43	0.71
Zn					1	-0.04	0.20	0.07	0.15	0.82*	-0.50
Cu						1	0.20	0.46	0.37	-0.29	0.68
Cd							1	0.61	0.42	0.20	0.00
Ba								1	0.93*	-0.29	0.64
Pb									1	-0.33	0.63
Mg										1	-0.82*
Fe											1

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

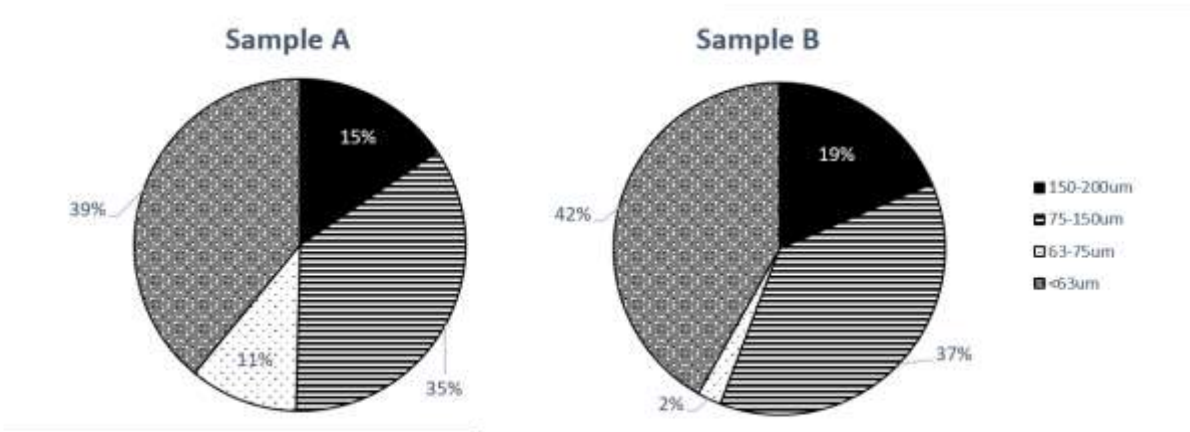

Figure 2. Pie chart showing the particle size distribution of Sample A and B

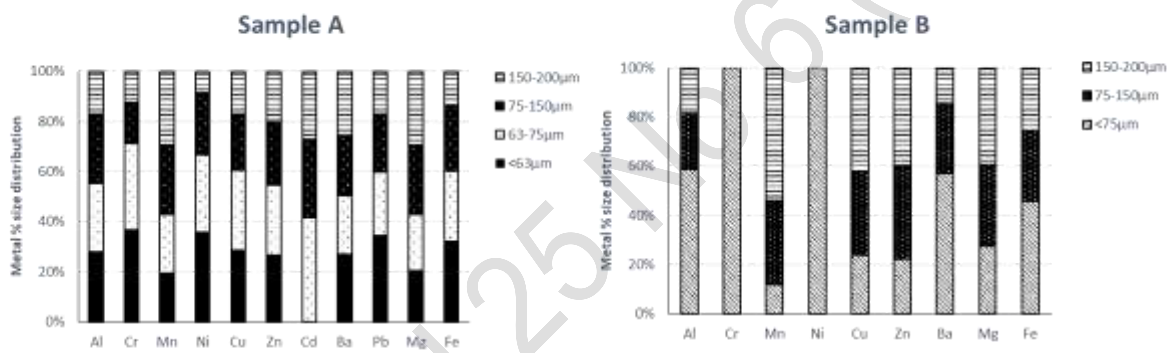

Figure 3. The distribution of heavy metals in different particle-size fractions of Sample A and B

Table 3. Concentrations of heavy metal in different size fractions

mg/kg	Sample A					Sample B			
	<63 µm	63-75 µm	75-150 µm	150-200 µm	<200 µm	<75 µm	75-150 µm	150-200 µm	<200 µm
Al	11000	10900	10900	6760	7740	4150	1610	1310	3240
Cr	69.0	65.0	31.0	23.2	33.0	140	BDL	BDL	0.710
Mn	706	847	1010	1060	782	1920	5620	8820	1720
Ni	72.6	63.0	50.9	17.3	43.4	37.3	BDL	BDL	6.24
Cu	129	145	102	76.2	81.9	116	166	204	65.3
Zn	967	1010	910	730	832	1110	1900	2000	836
Cd	BDL	1.130	0.850	0.740	0.190	BDL	BDL	BDL	BDL
Ba	446	390	398	422	420	85.9	43.4	22.0	72.6
Pb	100	72.9	67.2	49.0	55.1	BDL	BDL	BDL	BDL

Table 3 (cont'd). Concentrations of heavy metal in different size fractions

mg/kg	Sample A					Sample B			
	<63 µm	**63-75 µm**	**75-150 µm**	**150-200 µm**	**<200 µm**	**<75 µm**	**75-150 µm**	**150-200 µm**	**<200 µm**
Mg	2290	2490	3090	3280	2450	2890	3490	4180	2630
Fe	11500	10100	9290	4950	6070	4800	2940	2750	3190
Total	27300	26100	25900	17400	18500	15200	15800	19300	11800

BDL: Below detection limit

Table 4. Non-carcinogenic risks (HI) of heavy metals in indoor dust

	HQ_{ing}	**HQ_{inh}**	**HQ_{der}**	**HI**
Al	6.97E-03	2.60E-06	9.27E-05	7.06E-03
Cr	4.88E-03	1.82E-06	6.49E-05	4.94E-03
Mn	1.48E-03	5.50E-07	1.96E-05	1.50E-03
Ni	4.86E-04	1.81E-07	6.46E-06	4.92E-04
Zn	8.31E-03	3.10E-06	1.11E-04	8.43E-03
Cu	1.11E-04	4.12E-08	1.47E-06	1.12E-04
Cd	1.12E-05	4.16E-09	1.48E-07	1.13E-05
Ba	1.07E-03	4.00E-07	1.43E-05	1.09E-03
Pb	2.48E-03	9.25E-07	3.30E-05	2.52E-03
Mg	8.62E-05	3.21E-08	1.15E-06	8.74E-05
Fe	4.99E-03	1.86E-06	6.64E-05	5.06E-03

Conclusion

This study investigated the concentrations of selected heavy metals (Al, Cr, Mn, Ni, Zn, Cu, Cd, Ba, Pb, Mg, and Fe) from rural residential area of Simpang Renggam, Johor. Seven indoor dust samples (<200 µm) and two dust samples across size fractions <63 µm, 63-75 µm, 75-150 µm, and 150-200 µm were analyzed. The mean metal concentrations in bulk dust samples decreased in the following order: Fe> Al> Mg> Zn> Mn> Ba> Cu> Cr> Pb> Ni> Cd. For Fe and Al, the highest concentrations were found in the finest size fraction. For Mn, Mg, Cu, and Zn, the concentrations increased with increasing particle size, while no distinct trend could be observed for other metals. Human risk assessment based on <200 µm dust revealed HQ and HI values below than 1, indicating no non-carcinogenic risks from the exposure of these metals to the occupants. However, the enrichment of certain toxic metals in finer particle size suggested a likelihood of underestimating the exposure risk via inhalation. It should be kept in mind that the data set in this study is relatively small and therefore in future studies, investigation on the enrichment of a wider range of heavy metals in different particles size fractions of indoor dust collected from various environment would be more relevant for inhalation exposure assessment.

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