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Distribution, sources and health risks of heavy metals in indoor dust across China

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HIGHLIGHTS

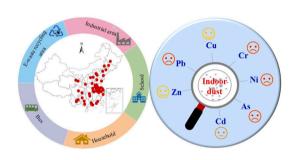
- The contaminations of heavy metals in Southern China were significantly higher than Northern China.
- The concentrations of Pb, Cu, and Cr during 2014–2021 were significantly lower than those in 1998–2013.
- Cd contamination level was the highest compared to other metals.
- Traffic emissions and decorative materials are the primary sources of heavy metals in indoor dust.
- More attention should be paid to the non-carcinogenic risk of Pb and As and cancer risk of Cr, Ni, and As.

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GRAPHICAL ABSTRACT



ABSTRACT

The potential effects of heavy metals on human health have attracted increasing attention as most people spend up to 90% of their time indoors. Human exposure to heavy metals in indoor dust have only been characterised for limited regions in China, and full-scale data for different functional areas are not available. Therefore, this review analysed the concentrations, contamination characteristics, and potential health risks of seven heavy metals (including zinc (Zn), lead (Pb), copper (Cu), chromium (Cr), nickel (Ni), arsenic (As), and cadmium (Cd)) in indoor dust at 3392 sampling sites in 55 cities across 27 provincial regions of China based on literature data. Results revealed that the median heavy metal concentrations in indoor dust throughout China decreased in the following order: Zn > Pb > Cu > Cr > Ni > As > Cd. Traffic emissions and decorative materials are the primary sources of heavy metal pollution in indoor dust. No considerable non-carcinogenic risk was found for Zn, Zn,

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1. Introduction

Indoor dust is a heterogeneous complex mixture of settled particulate matter originating from a range of indoor and outdoor sources (Al Hejami et al., 2020; Cao et al., 2017; Clarke et al., 2022; Hu et al., 2018), which is considered to be an important repository and transport carrier for a variety of environmental contaminants, including organic and inorganic pollutants (Cao et al., 2019a; Gillings et al., 2022; Mehmood et al., 2021c; Tan et al., 2016; Wang et al., 2021). Heavy metals (e.g. zinc (Zn), lead (Pb), copper (Cu), chromium (Cr), nickel (Ni), arsenic (As), and cadmium (Cd)) in indoor dust have gained attention worldwide because of their persistence, non-degradability, high toxicity, and adverse effects on human health (Nag and Cummins, 2022; Zhao et al., 2019; Zhou et al., 2021). Indoor heavy metal concentrations are generally higher than those of outdoor environments (Kurt-Karakus, 2012; Li et al., 2020; Zhao et al., 2022; Zhou et al., 2022). Furthermore, most people spend up to 90% of their time indoors, especially vulnerable infants, young children, and the elderly (Darus et al., 2012; Dong et al., 2019; Tan et al., 2016; Zhou et al., 2021). For example, children are more likely to be affected by indoor dust pollution than adults owing to their frequent hand to mouth activity, crawling behaviour, touching, and licking of toys (Cao et al., 2020; Dong et al., 2019, 2022; Ma et al.,

Heavy metals present in indoor environments can directly enter the human body via ingestion, inhalation, and dermal contact (Al Hejami et al., 2020; Doyi et al., 2020; Hahn et al., 2022; Mehmood et al., 2021a; Tan et al., 2016; Wang et al., 2010). Ingestion is the most important exposure route through which indoor dust affects human health (Cao et al., 2019b; Li et al., 2020; Lin et al., 2015; Wang et al., 2021). Heavy metals can act as co-factors or promoters of the central nervous system, heart, and other diseases and have adverse effects on cognitive development, intelligence quotient, and educational outcomes in children, even with low levels of Pb, Cr, As, and Cd exposure (Cao et al., 2020; Cheng et al., 2018; Doyi et al., 2019; Huang et al., 2012; Lanphear et al., 2018; Yoshinaga et al., 2014). It was reported that indoor dust was recognized as dominant source of Pb exposure for Japanese children, and the isotope ratios of Pb in their blood were similar to those in house dust (Takagi et al., 2011; Yoshinaga et al., 2014). Furthermore, the highest levels of blood Pb were found among children aged 2-3 years in Flin Flon, Canada, and the Pb concentration in household dust was associated with their blood Pb levels (Safruk et al., 2017). Another study indicated that children aged 1-2 years had the highest blood Pb levels in Sydney, Australia, and Pb and Cr exposure from indoor dust posed potentially adverse health risks for them (Doyi et al., 2019). Pb exposure from indoor dust is a major contributor to blood Pb poisoning in children in many developing countries (Shi and Wang, 2021). Therefore, heavy metal contamination in indoor dust and its adverse impacts on human health requires more attention.

There are regional and source differences in heavy metal contamination in indoor dust throughout China. For example, some previous reports have mainly focused on industrial cities, where strong industrial activities (such as smelting, mining activities and e-waste recycling) and high traffic density can lead to an increase in heavy metal levels in indoor dust (Cao et al., 2022; Li et al., 2020; Wang et al., 2021; Xu et al., 2015). In addition, various indoor environments can suffer from heavy metal pollution because of diverse indoor sources (Ajayi et al., 2022; Bao et al., 2019; Zhao et al., 2021; Zhou et al., 2022). In recent years, studies on heavy metals in indoor dust have focused on a single region or city worldwide (Alotaibi et al., 2022; Cao et al., 2020; Cheng et al., 2018; Dingle et al., 2021; Gul et al., 2022; Li et al., 2020; Wang et al., 2021). Meanwhile, a few of studies have compared the heavy metal contamination levels in indoor dust of several cities (Han et al., 2012; Shen et al., 2018; Zhao et al., 2020, 2021) or different functional areas of a city in China (He et al., 2017; Zhou et al., 2020). To our knowledge, a number of studies have examined the spatial distribution and contamination levels of heavy metals in soil (Shifaw, 2018; Sodango et al., 2018; Sun

et al., 2022; Wei and Yang, 2010; Yang et al., 2018) and road/street dust on a national scale in China (Hou et al., 2019; Wang et al., 2022). Several global-scale (Shi and Wang, 2021; Tan et al., 2016) and national survey (e.g. Canada (Rasmussen et al., 2013), Japan (Yoshinaga et al., 2014), and Germany (Seifert et al., 2000)) of heavy metal pollution in indoor dust have been conducted. However, for China, the contamination and human exposure to heavy metals in indoor dust have only been characterised or summarised for limited regions (Li et al., 2022; Liu et al., 2021), and full-scale data on heavy metal contamination assessment in indoor dust of different functional areas and their health risks across China are not available.

As few data is available for Hg and Mn in indoor dust, Zn, Pb, Cu, Cr, Ni, As, and Cd were selected for analysis in this study. We comprehensively evaluated the contamination characteristics and potential health risks of heavy metals in indoor dust of different functional areas on a national scale in China by analysing data from the published literature. The primary aims of this study were to: (1) evaluate the contamination levels and the differences in spatial and temporal distribution of Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust across China; (2) identify the contamination sources of Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust throughout China; and (3) analyse the spatial distributions of health risk and identify the primary contributors to efficaciously reduce human health risks in the future. Overall, this study helps in prioritising heavy metal pollution and its toxicity contribution to health implications of children and adults in China on a national scale.

2. Study methods and data sources

2.1. Data collection

In this study, we critically reviewed the concentrations of seven heavy metals (Zn, Pb, Cu, Cr, Ni, As, and Cd) in indoor dust from 27 provinces/autonomous regions/municipalities (including 55 cities and 3392 sampling sites) in China based on published literature. These seven heavy metals are listed as prioritised heavy metal pollutants by the United States Environmental Protection Agency (US EPA). All data were obtained through online electronic databases, such as Web of Science, PubMed, Google Scholar, Elsevier ScienceDirect, and two major databases of Chinese literature (including China National Knowledge Infrastructure and Wan-fang databases). Key words such as, 'heavy metal', 'metalloid', and 'trace elements' combined with 'indoor dust', 'house dust', 'household dust', and 'China' were used in the literature search. We chose literature that primarily focused on the pollution of Zn, Pb, Cu, Cr, Ni, As, and Cd and where the sample sites were dispersed throughout the whole country. Furthermore, we excluded reviews and articles with unclear heavy metal concentrations and articles that only reported bioavailable heavy metal concentrations in indoor dust. Sixty-three articles (sampling period between the years 1998-2021) on heavy metal concentrations in indoor dust across China were selected. The units of heavy metal concentrations from these relevant articles were standardised as mg•kg⁻¹. Dust samples were mainly collected from buses, households, offices, dormitories, hotels, schools, air conditioners, and ewaste workshops by using vacuum cleaners and clean plastic or polyethylene brushes. The analytical methods and tools used in these studies are all applicable and widely used by the scientific community. The concentrations, sample size, and sampling time of indoor dust for all selected sites are described in Table S1 of the Supplementary Information (SI).

2.2. Study methods

2.2.1. Geo-accumulation index (Igeo)

 I_{geo} is a quantitative indicator widely applied in the heavy metal pollution assessment in soil and dust (Hou et al., 2019; Ma et al., 2016; Mor et al., 2022), which can be calculated using the following equation:

$$I_{geo} = \log_2\left(\frac{C_n}{1.5B_n}\right) \tag{1}$$

where C_n represents the heavy metal concentration in indoor dust, and B_n is the background heavy metal concentration (Wang et al., 2021). The background values of soils (CNEMC, 1990) in every province were selected as geochemical background values because of limited background data available for indoor dust. The mean and background values of heavy metals in indoor dust for each investigated city are listed in Table S2. The I_{geo} values for heavy metals are divided into seven standard classes (Duan et al., 2020; Hou et al., 2019), which are summarised in Table S3.

2.2.2. Exposure assessment model

In this study, the health risk assessment model developed by the US EPA (US EPA, 2011) was used to evaluate human exposure to heavy metals in indoor dust. The average daily exposure dose (ADD) ($mg \cdot kg^{-1} \cdot day^{-1}$) of heavy metals through ingestion (ADD $_{ing}$), inhalation (ADD $_{inh}$), and dermal contact (ADD $_{dermal}$) can be estimated using Equations ((2)–(4)), respectively (Cheng et al., 2018; Hou et al., 2019; Tan et al., 2016).

$$ADD_{ing} = C \times \frac{\text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times 10^{-6}$$
 (2)

$$ADD_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT}$$
(3)

$$ADD_{dermal} = C \times \frac{\text{SL} \times \text{SA} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times 10^{-6}$$
 (4)

The average lifetime daily dose (LADD) (mg·kg⁻¹·day⁻¹) for Cr, Ni, As, and Cd via ingestion, inhalation, and dermal contact exposure pathways is used for cancer risk assessment. The formulas ((5)–(7)) for calculating LADD of Cr, Ni, As, and Cd in indoor dust through these three pathways are listed below (Doyi et al., 2019):

$$LADD_{ing} = C \times \frac{EF \times \left(\frac{ED_{child} \times IngR_{child}}{BW_{child}} + \frac{(ED_{adult} - ED_{child}) \times IngR_{adult}}{BW_{adult}}\right)}{AT_{ca}} \times 10^{-6}$$
(5)

$$LADD_{inh} = C \times \frac{EF \times ET \times ED}{PEF \times 24 \times AT_{ca}} \times 10^{3}$$
 (6)

$$LADD_{dermal} = C \times \frac{ABS_d \times EF \times DFS_{adj}}{AT_{ca}} \times 10^{-6}$$
(7)

where *C* is the concentration of heavy metals in the indoor dust. The detailed definitions and values of the parameters involved in the above mentioned equations are provided in Table S4.

2.2.3. Health risk assessment model

Hazard quotient (HQ) was used to evaluate the potential non-carcinogenic risk of each heavy metal in indoor dust. The HQ of ingestion (HQ $_{\rm ing}$), inhalation (HQ $_{\rm inh}$), and dermal contact (HQ $_{\rm dermal}$) pathways for each metal were then summed to yield a Hazard Index (HI) for representing the total risks (Wang et al., 2021; Zhang et al., 2019). The calculation ((8)–(11)) of HQ for each exposure pathway and HI are as follows:

$$HQ_{ing} = \frac{ADD_{ing}}{RfD_{ing}}$$
 (8)

$$HQ_{inh} = \frac{ADD_{inh}}{RfD_{inh}} \tag{9}$$

$$HQ_{dermal} = \frac{ADD_{dermal}}{\text{RfD}_{dermal}} \tag{10}$$

$$HI = HQ_{ing} + HQ_{inh} + HQ_{dermal}$$

$$\tag{11}$$

where RfD_{ing}, RfD_{inh}, and RfD_{dermal} are the daily intake reference doses (mg·kg⁻¹·day⁻¹) via ingestion, inhalation, and dermal contact, respectively. If the HQ and HI values are ≤ 1 , the non-carcinogenic effect is not significant. If the HQ and HI values are > 1, there is a high chance of non-carcinogenic effects. The RfD values for the three routes are provided in Table S5.

The incremental lifetime cancer risk (ILCR) was used to describe the carcinogenic risk of individual carcinogenic metal to humans during a lifetime via ingestion (ILCR $_{\rm ing}$), inhalation (ILCR $_{\rm inh}$), and dermal contact (ILCR $_{\rm dermal}$), and the calculations ((12)–(15)) are as follows:

$$ILCR_{ing} = LADD_{ing} \times SF_{ing}$$
 (12)

$$ILCR_{inh} = LADD_{inh} \times SF_{inh}$$
 (13)

$$ILCR_{dermal} = LADD_{dermal} \times SF_{dermal}$$
 (14)

$$ILCRs = ILCR_{ing} + ILCR_{inh} + ILCR_{dermal}$$
(15)

where SF_{ing}, SF_{inh}, and SF_{dermal} are the slope factors ((mg·kg $^{-1}\cdot day^{-1})^{-1}$) via ingestion, inhalation, and dermal contact, respectively. ILCRs is the sum of ILCR for the three pathways. The carcinogenic risk is acceptable or tolerable when ILCR and ILCRs are in the range of 1×10^{-6} to 1×10^{-4} . The SF values for the three routes are presented in Table S5.

2.3. Statistical analysis

In this review, Origin 2021 and ArcGIS 10.2 were used for diagraph analysis. SPSS Statistics 28 was used for all statistical tests and analyses, and the level of significance was set to p < 0.05. Spearman's correlation analysis and principal component analysis (PCA) were conducted to identify potential sources of heavy metals in indoor dust.

3. Results and discussion

3.1. Concentrations and profiles

3.1.1. Descriptive statistics of heavy metals in indoor dust

Heavy metal concentrations in indoor dust across China, based on the published literature, are shown in Table S1. The concentrations of Zn, Pb, Cu, Cr, Ni, As, and Cd ranged from 28.0 to 12500, 1.11-13600, 5.01–6290, 17.4–801, 2.50–1440, 1.06–210, and 0.07–699 mg kg⁻¹ respectively. The ratios of maximum to minimum for Zn, Pb, Cu, Cr, Ni, As, and Cd were 446, 12,300, 1260, 46.0, 575, 198, and 9990, respectively, indicating that the analysed heavy metals exhibited various degrees of enrichment in indoor dust throughout China. The median heavy metal concentrations in indoor dust decreased in the following order: Zn $(603 \text{ mg kg}^{-1}) > \text{Pb } (161 \text{ mg kg}^{-1}) > \text{Cu } (136 \text{ mg kg}^{-1}) > \text{Cr } (85.9 \text{ mg})$ kg^{-1}) > Ni (40.7 mg kg^{-1}) > As (15.6 mg kg^{-1}) > Cd (2.73 mg kg^{-1}) (Fig. S1). Comparatively, the concentrations of Pb (p < 0.01), Cu (p < 0.01) 0.05), and Cr (p < 0.01) in indoor dust during 2014–2021 were significantly lower than those in 1998-2013, while the concentration of As (p < 0.01) in indoor dust during 2014-2021 was significantly higher than that in 1998-2013. There were no significant differences in the concentrations of Zn, Ni, and Cd between 1998-2013 and 2014-2021 (Fig. 1 (a) and Table S6). As the suspended particulate matter in the atmosphere transferred to the room is one of the primary sources of indoor dust (Kurt-Karakus, 2012; Mehmood et al., 2021b; Wang et al., 2021), this significant temporal reduction in the concentrations of Pb, Cu, and Cr in indoor dust may be connected with the efforts of Chinese government to prevent air pollution in recent years (Guo et al., 2022; Qi et al., 2016) and the reductions in both air pollution and fossils fuel emission in response to community interventions in China during the COVID-19 pandemic (Mehmood et al., 2022a, 2022b).

Compared to literature data (Table S7), the median concentrations of

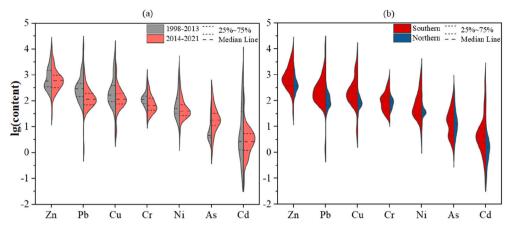


Fig. 1. Comparison of heavy metal concentrations in indoor dust (a) in China between 1998-2013 and 2014-2021 and (b) between Southern and Northern China.

Zn, Cu, Cr, Ni, and Cd in indoor dust on the nationwide scale in Canada were higher than those in this study, while the median concentrations of Pb and As were lower than those in this study (Rasmussen et al., 2013). Compared with Japan, the median concentrations of Pb, Cr, and Cd in indoor dust in China were higher, while the median concentrations of Zn, Cu, and Ni were lower (Yoshinaga et al., 2014). Additionally, the median concentrations of Zn, Pb, Cu, and Ni in indoor dust in the UK were higher than those in China, whereas the median concentration of Cd was lower than that in China (Turner and Simmonds, 2006). Compared with Australia, the median concentrations of Zn, Pb, Ni, and As were lower in this study, whereas the median concentrations of Cu and Cr were higher (Doyi et al., 2019). In summary, the concentrations of Zn, Cu, and Ni were at lower levels in this study, whereas the concentrations of Pb, Cr, As, and Cd were at a moderate level compared with other countries (Fig. S2).

3.1.2. Spatial distributions across China

A comparison of heavy metal concentrations in the southern and northern cities of China is exhibited in Fig. 1(b). The median concentrations of Zn, Pb, Cu, Cr, Ni, As, and Cd were higher in Southern China than those in Northern China. Actually, there were significant differences in the concentrations of Zn (p < 0.01), Pb (p < 0.01), Cu (p < 0.01) 0.01), and Cd (p < 0.01) in indoor dust between Southern and Northern China (Table S6). The spatial distributions of Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust throughout China were characterised using ArcGIS 10.2 to construct contamination maps with likely hotspots (Fig. 2). The results indicated that the heavy metal concentrations in indoor dust varied considerably among different geographic locations. High concentrations of Zn, Pb, Cu, Cr, Ni, and Cd were primarily observed in Southern China. For example, the maximum Zn and Cr concentrations in indoor dust were found in a smelting area in Qujing, Yunnan (Cao et al., 2020, 2022), and a college in Foshan, Guangdong (Cai et al., 2017), respectively. Pb and Cu concentrations in indoor dust in Wenling, Zhejiang were high, which might be attributed to family run workshops in remote areas that are involved in abandoned recycling for decades (Xu et al., 2015). The highest concentrations of Ni and Cd were observed in Hong Kong (Lau et al., 2014), which was related to two individual working areas (repair and dismantling) in formal e-waste recycling workshops and one informal e-waste recycling workshop that were chosen as the sampling sites. In contrast, low concentrations of Zn, Pb, Cr, Ni, and Cd were primarily observed in Northern China. Specifically, the lowest concentrations of Zn, Pb, Cr, Ni, and Cd were found in Suzhou, Anhui (Lin et al., 2015), Taiyuan, Shanxi (Zhao et al., 2021), Harbin, Heilongjiang (Gao et al., 2015), and Beijing (Cao et al., 2016), respectively. The lowest concentrations of Cu and As were observed in Taizhou, Jiangsu (Wu et al., 2016), and Wenling, Zhejiang (Liang, 2016), respectively.

3.1.3. Variations among different functional areas

The heavy metal concentration levels in indoor dust from five types of functional areas (e-waste recycling area, industrial area, school, household, and bus) are illustrated in Fig. 3. The distribution of different heavy metal concentrations in indoor dust varied considerably in different types of functional areas. The levels of Zn, Pb, Cu, Cr, Ni, and Cd were the highest in e-waste workshops, followed by those in industrial areas with the exception of Ni. These e-waste recycling areas are primarily distributed in Southern China. Smelting and e-waste recycling activities in Southern China might contribute prominently to heavy metal contamination in indoor dust (Zhu et al., 2012). The median concentration of As was the highest in industrial area, which may be influenced by industrial discharges or ore-related activities, such as ore mining and smelting (Cao et al., 2020, 2022). The concentrations of these seven heavy metals in school dust were comparable to those in household dust, and bus dust presented the lowest levels of heavy metals among the five types of functional areas.

The composition profiles of heavy metals exhibited clear differences among the five types of functional areas. Specifically based on the median concentrations, Zn was the dominant component in industrial areas, schools, households, and bus dust, accounting for more than 50% of the total heavy metals, contributing more than the e-waste recycling area (Fig. S3). The percentage proportions of Cu and Ni in the e-waste workshop were higher than those in the other four functional areas, accounting for 25.3% and 5.75% of the total heavy metals, respectively. The contributions of Pb (23.7%) and Cd (1.01%) in the industrial area were higher than those in the other four types of functional areas. The percentage proportion of Cr (10.6%) was the highest in household dust, followed by that in school dust. The contribution of As (4.12%) in the industrial area was the highest compared with the other four types of functional areas.

3.2. Contamination level assessment

As exhibited in Table S2, the I_{geo} values for Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust across China were in the range of 0.82–5.78, 0.41–6.79, -2.52–6.55, -1.51–2.71, -1.18–2.60, -2.48–3.97, and 0.15–9.36, respectively. Based on the standard classes of I_{geo} , the contamination distributions of heavy metals in indoor dust on a national scale are shown in Fig. 4. In detail, the I_{geo} values of Cu, Cr, Ni, and As in 6.12%, 37.2%, 52.4%, and 50% of the selected cities, respectively, were found to be lower than 0, such as, Cu in Zhengzhou, Taizhou, and Changde; Cr in Harbin, Changchun, Shenyang, Lanzhou, Qingdao, and Hefei; Ni in Harbin, Changchun, Dalian, Shijiazhuang, Beijing, and Tianjin; and As in Harbin, Taiyuan, Jinan, Hefei, and Shanghai, suggesting that indoor dust in these cities was not contaminated by the corresponding metals. Apart of these cities, the indoor dust in other

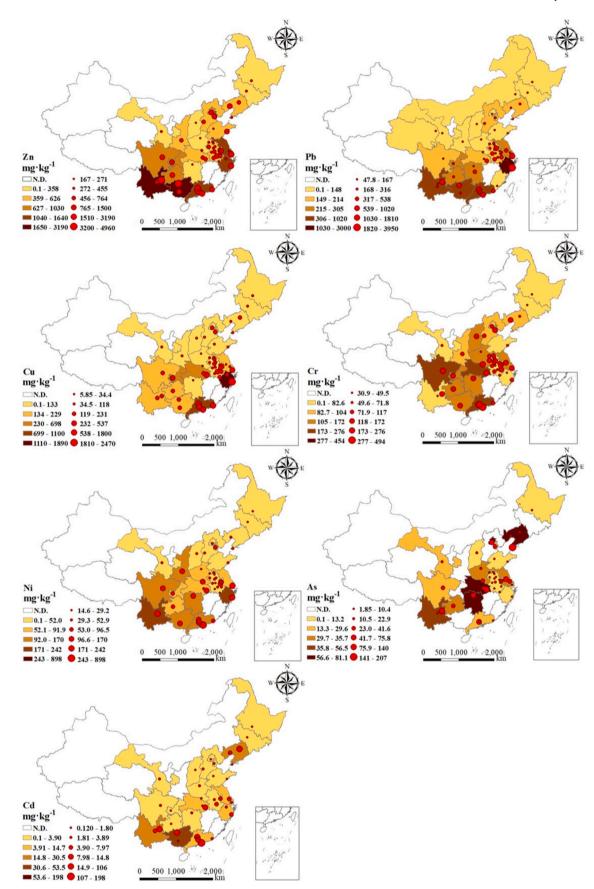


Fig. 2. Spatial distributions of Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust throughout China (Red circles represent heavy metal concentration levels in different cities, and different colours indicate mean heavy metal concentration levels in different provinces). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

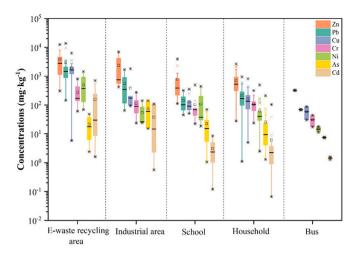


Fig. 3. Heavy metal levels in indoor dust of different functional areas.

cities was contaminated by the seven heavy metals to varying degrees. The maximum $I_{\rm geo}$ values for Zn (5.78), Cr (2.71), and Ni (2.60) were observed in Foshan, Guangdong; the maximum $I_{\rm geo}$ values of Pb (6.79) and Cu (6.55) were observed in Wenling, Zhejiang; and the maximum $I_{\rm geo}$ values of As (3.97) and Cd (9.36) were observed in Dalian, Liaoning, and Qingyuan, Guangdong, respectively. This exhibited that the indoor dust in these cities was moderately to extremely contaminated by the corresponding metals. Comparatively lower levels of contamination were found in Northern China, including Harbin, Changchun, Jarud Banner, Beijing, and Taiyuan.

The mean $I_{\rm geo}$ values for Zn, Pb, Cu, Cr, Ni, As, and Cd in indoor dust across China were 2.61, 2.48, 2.09, 0.08, 0.34, -0.07, and 4.36, respectively. The results indicated that Zn, Pb, and Cu, Cr and Ni, and As fell into the categories of 'moderately to strongly contaminated', 'uncontaminated to moderately contaminated', and 'practically uncontaminated', respectively. Only Cd was in the category of 'strongly polluted to extremely contaminated' in China. Therefore, it is necessary to pay more attention to Cd contamination in indoor dust. Statistically, the contamination levels of Zn (p < 0.05), Pb (p < 0.01), Cu (p < 0.05), Cr (p < 0.05), Ni (p < 0.01), and Cd (p < 0.05) in indoor dust in Southern China were significantly higher than those in Northern China (Table S6), while there were no significant differences in contamination levels of As (p > 0.05) between Southern China and Northern China.

3.3. Source identification

Currently, the sources of heavy metals in indoor dust are identified based on PCA (Duan et al., 2020; Isley et al., 2022; Qin et al., 2015; Wang et al., 2021), enrichment factors (Cheng et al., 2018; Ma et al., 2020; Xu et al., 2015), and positive matrix factorisation (Huang et al., 2022; Wang et al., 2021). Heavy metal contamination in indoor dust is influenced by the combined effects of external and internal sources over a long period of time (Zhao et al., 2022). Outdoor sources include traffic emissions, industrial activities, smelting, mining activities, local soil, and road dust. Indoor sources primarily include smoking, cooking, fuel combustion, building structure, and decorative materials (such as interior decorations, floor and coating materials, carpets, and paints) (Isley et al., 2022; Li et al., 2020; Liu et al., 2021; Shi and Wang, 2021; Zhou et al., 2022).

The contribution of different sources to the total contamination based on published sources of heavy metals in indoor dust is illustrated in Fig. 5(a), which exhibits that the statistical number of cities emerged a certain source divided by the total number of surveyed cities. For outdoor sources, traffic emissions, industrial activities, local soil, road dust, mining activities, and smelting contributed to heavy metal pollution in indoor dust in 49.4%, 30.3%, 12.4%, 9.0%, 7.9%, and 5.6% of the

surveyed cities, respectively. This indicates that traffic is the most important source of heavy metals in indoor dust, followed by industrial activities. Pb and As levels in household dust are associated with traffic density near home because traffic exhaust emissions lead to the enrichment of Pb in dust (Lin et al., 2016; Zhao et al., 2022). In addition, traffic from motor vehicles (including tire abrasion, vehicle brake linings, friction of vehicle metallic parts, and roads) leads to Zn, Ni, and Cd pollution (Cheng et al., 2018; Ma et al., 2020). Heavy metal pollution in Xi'an, Qingdao, Tongling, and other cities may be primarily derived from traffic emissions (Li et al., 2020; Zhao et al., 2019). Industrial activities, including e-waste recycling, are the dominant sources of heavy metal pollution in indoor dust of typical industrial cities, such as Wenling, Daye, Guiyu, Qingyuan, and Hong Kong (Lau et al., 2014; Wang et al., 2021; Xu et al., 2015; Yu et al., 2019; Zhu et al., 2012). Furthermore, Cd and Cr emissions predominantly originate from various industrial activities (including chemical engineering, electroplating, printing and dyeing, and waste incineration) (He et al., 2017; Zhao et al., 2019).

Among the indoor sources, decorative materials were the dominant contributor to heavy metal pollution in indoor dust in 43.8% of the surveyed cities. Wall paint and coating materials may lead to the accumulation of Zn, Pb, Cu, and Cd (Bao et al., 2019; Cheng et al., 2018; Duan et al., 2020; Li et al., 2016; Lin et al., 2016). Moreover, the colour of the wall paint is the most crucial factor for heavy metal pollution in indoor dust. For example, yellow paint is extensively associated with Zn, Cu, Pb, and Cd contamination, whereas purple and green paint have been reported to contain high levels of Zn, Pb, and Cu (He et al., 2017; Kurt-Karakus, 2012; Li et al., 2016). In addition, smoking and fuel combustion contributed to heavy metal contamination in indoor dust in more than 20% of the surveyed cities, such as Chengdu, Dalian, Taiyuan, and Lanzhou (Cheng et al., 2018; Wang et al., 2021; Zhao et al., 2021).

In this study, correlation analysis and PCA were applied to identify potential sources of heavy metals in indoor dust. Spearman's correlation analysis of the seven heavy metals in indoor dust across China is presented in Table S8. There were significant positive correlations between Pb–Cu–Cd (p < 0.01). In addition, significant correlations were observed between Zn and Cr (r = 0.354; p < 0.05), Zn and Ni (r = 0.619; p < 0.01), and Cr and Ni (r = 0.608; p < 0.01). Strong correlations between heavy metal concentrations in samples might reflect heavy metals originating from the same sources (Li et al., 2020; Mehmood et al., 2021a; Zhao et al., 2022; Zhou et al., 2020). This indicates that Pb, Cu, and Cd, Zn, Cr, and Ni may have a common source. Two principal components were extracted according to the PCA results presented in Fig. 5(b) and Table S9. The cumulative contribution rate reached 63.1%, which indicated that most information related to the sources of seven heavy metals in indoor dust across China was comprised. The first component (PC1) accounted for 45.6% of the total variance and was dominated by Pb, Cu, and Cd, which might have been derived primarily from traffic emissions and industrial activities. For instance, Pb emissions can occur from the use of leaded gasoline (Lin et al., 2015). Cu and Cd pollution results from gasoline use, oil leakage, tire abrasion, and corrosion of car bodies, as well as industrial activities, including smelting and mining (Cheng et al., 2018; Hou et al., 2019). The second component (PC2) with a variance contribution of 17.5%, demonstrated high loadings of Zn, Cr, and Ni, which might be attributed to indoor sources, particularly decorative materials. Shanghai, Wengling, Daye, Hong Kong, Qujing, Foshan, Hechi, Qingyuan, and Guiyu represented less scattered PCA profiles and were controlled primarily by PC1. Heavy metals in most cities were located in the centre of the plot, with PC1 and PC2 making important contributions (Fig. 5(b)), indicating relatively more complex sources of heavy metals compared with those of Shanghai, Wenling, Daye, Hong Kong, Qujing, Foshan, Hechi, Qingyuan, and Guiyu.

All source apportionment results obtained using the two independent methods were consistent. The results of PCA and correlation analysis were in accordance with the findings of the percentage of different sources of heavy metals in indoor dust on a national scale, indicating

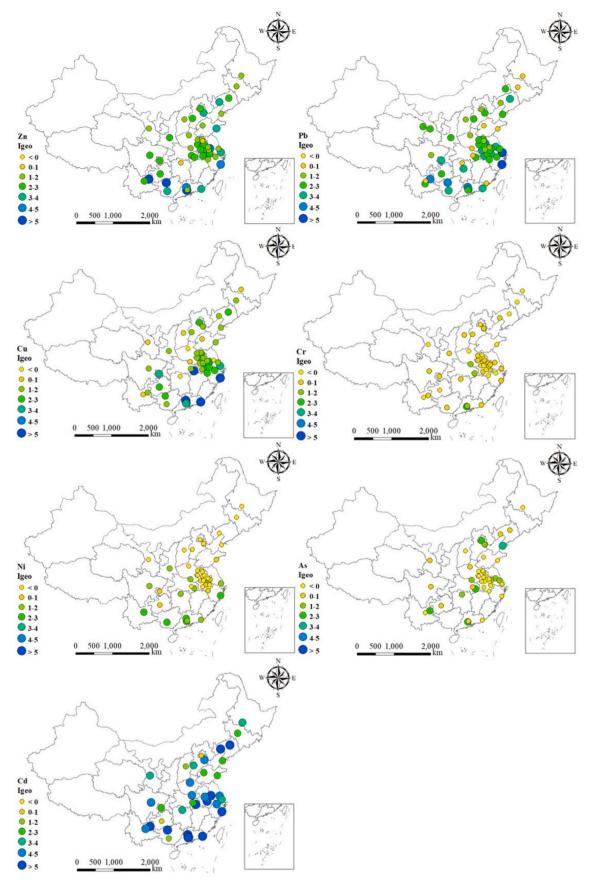


Fig. 4. Heavy metal contamination levels in indoor dust of different cities in China.

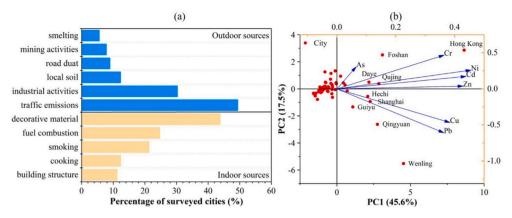


Fig. 5. (a) Major sources of heavy metal contamination in surveyed cities, and (b) principal component analysis (PCA) results based on heavy metal concentrations in indoor dust across China.

that traffic emissions, industrial activities, and decorative materials are the dominant contributors to heavy metal pollution in indoor dust across China. Therefore, it is crucial to reduce traffic, industrial emissions, and decorative material pollution to alleviate heavy metal pollution in indoor dust.

3.4. Human exposure and health risk assessment

3.4.1. ADD and non-cancer risk assessment

In this study, the ADD values of the seven heavy metals in indoor dust across China via oral ingestion, dermal contact and inhalation for both children and adults were evaluated. As exhibited in Tables S10–16, the ADD values of the seven heavy metals for both children and adults via the three exposure pathways decreased in the following order: ADD $_{\rm ing} > {\rm ADD}_{\rm dermal} > {\rm ADD}_{\rm inh},$ indicating that ingestion is the most dominant exposure pathway, which is consistent with some previous studies on heavy metals in urban street/road dust across China (Hou et al., 2019; Wang et al., 2022). Similar results were obtained for indoor dust in other countries, such as Australia (Doyi et al., 2019), Serbia (Buljovcic et al., 2022), Nepal (Yadav et al., 2019), and Turkey (Gul et al., 2022). Additionally, children were more vulnerable to heavy metal exposure compared with adults.

The non-cancer risks levels (including HQ and HI) posed by the seven heavy metals in indoor dust for both children and adults are presented in Tables S17–23. Specifically, the HQ values of these metals for both children and adults via oral ingestion, dermal contact, and inhalation

ranked in the following order: $HQ_{ing} > \, HQ_{dermal} \, > \, HQ_{inh}.$ This is consistent with the abovementioned ADD results. As exhibited in Fig. 6 (a) and Table S6, the HI values of the seven heavy metals (Zn, Pb, Cu, Cr, Ni, As, and Cd (p < 0.01)) for children were significantly higher than those for adults, indicating that children were more susceptible to heavy metal pollution in indoor dust than adults. The HI values of Pb, Cr, and As were higher than those of Zn, Cu, Ni, and Cd for both children and adults. For children, the HI values of Zn, Cu, Cr, Ni, and Cd were lower than 1 (the safe level) in all cities, whereas the HI values of Pb in Southern China (such as Wenling, Guangzhou, Qingyuan, Hong Kong, and Qujing) and As in Dalian, Beijing, Daye, Changde, and Qujing were higher than the safe level (Fig. S5). Thus, more attention should be paid to Pb and As exposure in these areas, especially in Southern China. The HI values of Zn, Pb, Cu, Cr, Ni, As, and Cd for adults were lower than the safe level in all cities, suggesting that there is no non-cancer risk from the seven heavy metals in indoor dust for adults. The non-cancer risks posed by Zn (p < 0.05), Pb (p < 0.01), Cu (p < 0.01), Cr (p < 0.05), Ni (p < 0.05), Vi (p < 0.05< 0.01), and Cd (p < 0.01) in Southern China (Guangzhou, Qingyuan, Hong Kong, and Daye) were significantly higher than in Northern China for both children and adults (Fig. S5 and Table S6), which is consistent with the above described spatial distribution of heavy metal contamination in indoor dust. Accordingly, effective measures in Southern China should be prioritised for controlling Pb and As pollution. Moreover, children are the key population that needs to be protected, and the potential non-cancer risks to children via ingestion of indoor dust cannot be neglected.

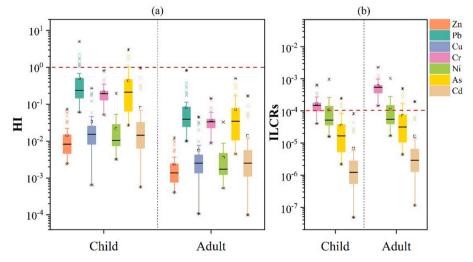


Fig. 6. (a) Hazard Index (HI) and (b) lifetime carcinogenic risk (ILCR) of selected, heavy metals for child and adult.

3.4.2. LADD and cancer risk assessment

The LADD values of Cr, Ni, As, and Cd in indoor dust across China via oral ingestion, dermal contact, and inhalation for both children and adults are provided in Tables S24–27. The LADD $_{ing}$ values of these four heavy metals for children and adults were the highest, followed by LADD $_{inh}$ and LADD $_{dermal}$, which was slightly different from the ADD of heavy metals.

Based on the calculation of the carcinogenic risk (cancer risk; ILCR) of exposure to Cr, Ni, As, and Cd in indoor dust across China (Tables S28-31), the total ILCR values of these heavy metals for both children and adults decreased in the following order: Cr > Ni > As > Cd (Fig. 6(b)). Moreover, the total ILCR values of Cr (p < 0.01), As (p < 0.01) 0.05), and Cd (p < 0.01) were significantly higher in adults than in children (Table S6). For both children and adults, the ILCR values of Cr in most surveyed cities were greater than 1×10^{-4} (unacceptable level), whereas the ILCR values of Ni, As, and Cd in most surveyed cities ranged from 1×10^{-6} to 1×10^{-4} (acceptable level), indicating that Cr in indoor dust poses a higher cancer risk for both children and adults in most cities in China. Moreover, the proportional ILCR values of Cr in indoor dust through inhalation for both children and adults were considerably higher than those for the other two exposure pathways (Fig. S6), suggesting that the inhalation of Cr was the dominant exposure pathway, followed by ingestion and dermal pathways. The ILCR distributions of Cr, Ni, As, and Cd in indoor dust for both children and adults on a national scale are illustrated in Fig. S7. For children, the total ILCR values of Cr, Ni, As and Cd in indoor dust of 55 cities across China ranged from 4.02×10^{-5} (Harbin, Heilongjiang) to 6.43×10^{-4} (Foshan, Guangdong), 1.58×10^{-5} (Harbin, Heilongjiang) to 9.72×10^{-4} (Hong Kong), 2.20×10^{-6} (Wenling, Zhejiang) to 2.47×10^{-4} (Dalian, Liaoning), and 4.84×10^{-8} (Beijing) to 8.19×10^{-5} (Hong Kong), respectively, implying that the ILCR distributions for heavy metals in indoor dust varied remarkably among the studied cities. In particular, the cancer risks of Cr and Ni in children (Guangzhou, Foshan, Nanning, and Hong Kong) arouses much concern in Southern China. The total ILCRs distribution of the four heavy metals in adults was similar to that in children.

3.4.3. Significance of exposure to heavy metals in indoor dust

Compared with other environmental matrices (Table S32), we observed that the mean HI values of Zn, Pb, Cu, Cr, Ni, and Cd in road dust across China for children and adults were lower than those in this study, except for the HI values of Cr for adults (Wang et al., 2022). The mean HI values of Zn, Pb, Cu, and Cd for both children and adults in this study were higher than those in urban street dust across China (Hou et al., 2019). Furthermore, the mean non-carcinogenic risk values of Zn, Pb, and Cu via inhalation in PM₁₀ in traffic environments in China were considerably higher than those in this study (Chen et al., 2015). The HI values of Zn and Cr for both children and adults and the HI values of Pb, Cu, and Ni for children in Australia were higher than those in this study; in particular, the HI values of Pb and Cr for children exceeded acceptable limits (Doyi et al., 2019), whereas the HI value of As for children and adults was higher in this study. Based on the limited literature data, the total non-carcinogenic risk of Zn, Pb, Cu, Cr, Ni, and Cd analysed in this study was at a moderate level, whereas the HI value of As was at a higher level. Indoor dust is probably the primary contributor to exposure to heavy metals, and research on indoor dust exposure should be conducted in the future. Moreover, greater attention should be paid to the non-carcinogenic risks posed by Pb and As in indoor dust.

3.5. Limitations and uncertainties

This study had several limitations. The published scientific literature and collected data on heavy metals in indoor dust across China are limited. There are no data on heavy metals in indoor dust from several provinces and autonomous regions, such as Ningxia, Tibet, Xinjiang, and Jiangxi. Thus, this study could not comprehensively analyse the overall heavy metal contamination status of indoor dust on a national scale.

Further studies are required to investigate heavy metal pollution in indoor dust in various regions of China, specifically in Northwest China. Additionally, the particle size of indoor dust from different cities in this study varied greatly, ranging from $<\!500~\mu m$ to $<\!20~\mu m$, and the distributions of heavy metal concentrations varied with different particle sizes of indoor dust, which influences the accuracy of health risk assessment of human exposure to heavy metals in indoor dust (Cao et al., 2015; Doyi et al., 2020). Therefore, it is essential to study heavy metal concentrations in dust samples with uniform particle size that provide more realistic contrasting results. Furthermore, other factors, such as temporal and seasonal variation of sampling, regional differences, house characteristics and decoration materials, and industrial activities, affect the concentration level assessment and source identification of heavy metal contamination in indoor dust on a national scale.

4. Conclusion

This study provides comprehensive knowledge on the distribution, contamination levels, sources, and health risks of heavy metals in indoor dust across 27 provincial regions of China. The concentrations of Pb, Cu, and Cr in indoor dust during 2014-2021 were significantly lower than those in 1998-2013. The heavy metal concentrations in indoor dust from e-waste workshops and industrial areas were higher than those in schools, households, and buses. In general, Zn, Pb, Cu, Cr, Ni, and Cd contamination in Southern China was higher than that in Northern China. Only Cd displayed the category of 'strongly polluted to extremely contaminated' in China. Compared with other foreign countries, the concentrations of Zn, Cu, and Ni in indoor dust across China were low, while Pb, Cr, As, and Cd were at a moderate level. The HI values of Pb and As for children exceeded the safe level (1) primarily in cities in Southern China, indicating a potential non-carcinogenic risk. The carcinogenic risks of Cr, Ni, and As for both children and adults in some surveyed cities, especially in Southern China, exceeded the unacceptable level (1 \times 10⁻⁴). Therefore, more attention should be paid to the noncancer risks and cancer risks posed by Pb and As and Cr, Ni, and As, respectively. In conclusion, these findings provide insights on the distribution of heavy metals in indoor dust and prioritise their pollution and toxicity contributions, especially identifying their primary contributors and regions with considerable heavy metal pollution in indoor dust across China, which is valuable for potential interventions aimed at reducing exposures and health risks associated with heavy metals in the future.

Credit author statement

Mengmeng Wang: Methodology, Writing – original draft, Writing – review & editing, Data curation, Visualization. Yinyi Lv: Conceptualization, Data curation. Xinyan Lv: Conceptualization, Data curation. Qianhan Wang: Conceptualization, Data curation. Yiyi Li: Visualization. Ping Lu: Visualization. Hao Yu: Writing – review & editing. Pengkun Wei: Writing – review & editing. Zhiguo Cao: Conceptualization, Supervision, Funding acquisition, Writing – original draft, Writing – review & editing. Taicheng An: Conceptualization, Supervision, Funding acquisition, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.137595.

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