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Comprehensive assessment of soil and dust heavy metal(loid)s exposure scenarios at residential playgrounds in Beijing, China

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Highlights

- Health risk of metal(loid)s exposure at residential playgrounds was assessed
- A positive correlation was observed between the levels of As (Ni) and Mn in soil
- Significant risk contribution prioritized As and Ni as control metal(loid)s
- Slight CRs and negligible non-carcinogenic risk were posed to children
- Significant health risk concentrated in suburbs rather than in urban areas

Abstract

Small playgrounds situated within residential communities are popular recreational areas. However, heavy metal(loid)s (HMs) in soil or equipment dust may pose a public health risk. This study provides a comprehensive assessment of the health risk associated with HMs exposure at residential playgrounds in cities, a field that has not been thoroughly investigated previously. 70 soil and 70 equipment dust samples were collected from 30 urban and 40 suburban playgrounds in Beijing. Results indicated significant enrichment of Cu, As, and Ni in the soil with Enrichment Factors (EFs) >5 from both anthropogenic and lithogenic sources. Correlation analyses showed that the levels of Be, Cr, Mn, Co, Ni in soil and Be, Mn, As, Cd in dust were positively correlated with the distance to the nearest highway, with *p-values* <0.01. Enrichment and correlation analyses contributed to a better understanding of the sources and transport pathways of HMs in urban environment. Based on a site-specific Conceptual Site Model (CSM), the carcinogenic risks (CRs) and Hazard Quotients (HQs) were quantified for residents as the ratio of HMs exposure to reference doses. Risk assessment indicated the mean predicted CR for children and adults exposed to soil was 3.75×10^{-6} and 5.29×10^{-6}, respectively, while that at dust exposure scenarios was lower, at 2.47×10^{-6} and 3.49×10^{-6}, respectively, all of which were at the upper end of U.S. EPA's acceptable criteria of 1×10^{-6} to 1×10^{-4}. Among the HMs, As and Ni were identified as the priority control contaminants due to significant contribution to CRs. Futhermore, the spatial distribution revealed an increasing trend in health risk from the urban center to the suburbs. This study emphasizes the need for effective measures to mitigate potential health risk and enhance the safety of recreational areas, particularly for susceptible individuals.

Keywords: Residential playgrounds, heavy metal(loid)s, equipment dust, health risk assessment, risk management

1 Introduction

Human health risk associated with urban soil or dust HMs exposure has attracted significant attention from scientists and public health practitioners. Exposure to HMs is known to cause DNA damage, cell death, tissue injury, organ dysfunction, behavioral disorders, mental retardation, and neurological disorders(Jin et al., 2019; Zhang et al., 2022; Zheng et al., 2010). Chronic exposure to As, Cd, and Ni, which are classified as group 1 carcinogens by the International Agency for Research on Cancer, also increases the risk of lung and respiratory tract cancer(Hou et al., 2019; Leung et al., 2008; Lu et al., 2015; Zheng et al., 2010). Rapid industrialization and urbanization in China have resulted in the widespread accumulation of HMs in urban soil, dust, and air, posing a significant health risk to residents. A national soil quality survey of China revealed that 16.1% of soil samples exceeded environmental quality standards (GB15618-1995), with key contaminants being Cd, Ni, and As(Ministry of Environmental Protection et al., 2014).

Based on bibliometric analysis of the Web of Science database, the number of publications related to environmental behavior and risk of HMs in dust or soil has increased rapidly in recent years. The research trend is depicted in Fig. 1. The publications relate to several key metal(loid)s in the following order of prevalence: Pb >Cd >Ni >Cr >As (Fig. 1d). The bibliometric analysis also indicates that the research mainly focused on the following topics: 1) Spatial distribution and source apportionment(Liu et al. 2021; Yadav et al. 2018; Dong et al. 2022); 2) Spatial migration and species transformation(Wiseman et al. 2018); 3) Environmental risk quantification as well as practical risk management (Al-Swadi et al. 2022). This finding underscores the significance of conducting additional research to comprehend the health risk of children and adults from chronic exposure to HMs in specific scenarios, such as residential playgrounds.

Numerous studies have focused on the source, transport, spatial distribution, and potential health risk associated with HMs in urban environments (Fig. 1c)(Adimalla et al., 2020; Hou et al., 2019; Hu et al., 2011; Tashakor et al., 2021; Tepanosyan et al., 2017; Xie et al., 2017). However, previous research mainly concetrated on the identification of contaminants and their sources. Limited information is available regarding the health risk posed by chronic exposure to HMs in specific scenarios, such as residential playgrounds where exposure may occur frequently but can be easily overlooked. Moreover, the relationship between environmental, social, and economic factors and health risk remains unclear.

In terms of health risk assessment, the population category is regarded as a crucial factor that significantly affects the ultimate risk characterization. Attention has increasingly been focused on the risk to children, who are at a considerably higher risk for negative health impacts than adults (Chen et al., 2014; Lin et al., 2016; Tao et al., 2012). It is due to their low tolerance as well as high exposure dose to soil or dust through hand to mouth, ingestion and other exposure pathways (Fig. 1b)(Wang et al., 2022).

(Fig. 1)

Many previous studies have focused on the relationship between HMs accumulation and environmental variables of hypothetical sources in residential communities. Based on these studies, risk management measures are designed and put into practice. However, conventional risk management strategies often encounter many challenges, including various levels of adverse impacts on the environment, society and human health at different levels. As a result, the implementation of risk management often fails to meet expectations. Feasible and effective approaches to mitigate potential adverse health effects on both children and adults in residential areas are aimed to be developed in this study.

This study aims to assess the potential health risk posed by exposure to HMs at community playgrounds. The main objectives are 1) to determine the concentrations and distribution of HMs in soil and equipment surface dust at residential playgrounds; 2) to develop a site-specific CSM for assessing health risk associated with long-term exposure to HMs; 3) to identify priority control contaminants and hotspots. By providing a comprehensive understanding of HMs exposure at residential playgrounds, this study may contribute valuable insights for future decision-making in risk management, ultimately leading to a cleaner and safer recreational environment.

2 Methodology

2.1 Case study

As the capital city, political center and cultural center of China, Beijing (E115.7º-

117.4°, N39.4°-41.6°) consists of 16 administrative districts, including 6 central districts located in the main urban areas, and the others belonging to suburban areas. It has typical semiarid monsoon climate with an average annual precipitation of 483.9 mm and average temperature range of 14.4-38°C. As of 2022, the total population has reached 21.84 million, with an annual Gross Domestic Product (GDP) of USD 601.52 billion. Rapid development in infrastructure and consequent increase in vehicle ownership have had a negative impact on soil quality due to road pavement, fuel leakage from car accidents, exhaust emissions, tire abrasion, runoff from highways, and other pollution sources.

2.2 Sample collection

The sampling sites covered the entirety of Beijing City, including 6 urban districts (such as Haidian, Chaoyang) and 10 suburban districts (such as Mentougou, Tongzhou). In urban districts, the sampling sites were mainly located in residential communities, while those in suburban districts were situated in residential communities, small towns and tiny villages. A total of 70 residential playgrounds, 30 in urban areas and 40 in suburban areas, were selected for sampling. Detailed spatial distribution and brief descriptions associated with urban and suburban sites were shown in Fig. 2 and Table S1. Sampling density was adjusted in urban and suburban areas, considering constraints such as limited funding, manpower, and the extensive study area. The sampling sites were arranged to align with the corresponding community densities. As shown in Fig. 2, the density of residential communities in urban areas ranged from 2.66 to 22.96 communities per square kilometer, whereas in suburban areas, it varied from 0.03 to 0.70 communities per square kilometer. Tongzhou District was chosen for denser sampling due to its higher residential community density and larger number of residential playgrounds compared to other suburbs. At each site, approximately 1 kg of topsoil (0-10 cm), consisting of five sub-samples, was collected from the ground surface using a woody spade. Sub-samples were distributed in a "plum-blossom-layout" at the center and four corners of the playground. Around 5 g of dust was swept from the surface of playground equipment using a polyethylene brush and then collected in a tight-sealed quartz glass vial. The tools were washed using deionized water before the next sampling to prevent cross-contamination. The GPS coordinates and site description were recorded using a portable GPS tracker (Juno SC, Trimble, USA).

Environmental variables, including vegetation coverage ratio, distance to the closest construction sites, factories, highways, and streets, were also recorded simultaneously. The quality control measures in the processes of conservation, transportation were implemented based on *the Technical Specification for Soil Environmental Monitoring (HJ/T 166-2004)* (MEE, China)(Chinese Ministry of Ecology and Environment 2004).

(Fig. 2)

2.3 Pretreatment and chemical analysis

The original soil and dust samples were stored at <4ºC prior to next processing. Firstly, stone, plant residue, and other debris were manually removed from soil and dust. The samples were subsequently dried at 105ºC in an oven until a consistent mass was achieved. Secondly, the soil samples were ground in a porcelain mortar before passed through a 100 mesh sieve. Approximately 0.25-0.50 g of soil or dust was then digested with 9 mL of concentrated HNO_3 and 3 mL of concentrated HCl reagents in a 50 mL polytetrafluoroethylene digestion vessel, following the technical requirement of U.S. EPA Method 3052 (the United States Environmental protection agency 1996). The polytetrafluoroethylene digestion vessels were placed in a fume hood before microwave digestion (CEM Mars 6, U.S. EPA). The digestion solution was diluted to 50 mL with 2% concentrated HNO_3, passed through 0.25 μm filters, and preserved at < 4ºC before laboratory analysis. To avoid glassware cross-contamination, the vessels were immersed in concentrated HNO_3 and subsequently rinsed at least three times with DI water.

The concentrations of 11 elements (Be, Cr, Mn, Co, Ni, Cu, As, Se, Cd, Sb, Fe) in each soil or dust solution were determined using an ICP-MS (inductively coupled plasma mass spectrometer, XSERIES 2, Thermo Fisher Scientific, USA). Quality Assurance and Quality Control measures were implemented in accordance with U.S. EPA Method 6020A (SW-846): Inductively Coupled Plasma-Mass Spectrometry(the U.S. Environmental Protection Agency 1998), which included the use of reagent blanks, certified references, triplicate analyses, internal standard, and others. Calibration standard solutions were prepared by diluting 100 mg/L to achieve concentrations ranging from 0-100 μg/L for each element. Prior to analysis, a linear calibration curve with a regression coefficient >0.999 was established. The instrument was monitored for stability and consistency every 20 samples.

2.4 Contamination evaluation

2.4.1 Geoaccumulation Index

Geoaccumulation index (I_{ge}) was used to evaluate soil contamination(Chen et al., 2015; Muller 1969). The coefficient of 1.5 was used to offset the variations of background levels of B_n in the natural environment. GIs could be calculated using the following formula:

$$I_{ge} = log_2(\frac{C_n}{1.5B_n}) \tag{2-1}$$

where C_n represents the concentration of "n" HM, and B_n means the background value of "n" HM. The levels of soil contamination were classified into seven different categories based on the geoaccumulation values (Table S2)(Trujillo-González et al., 2016).

2.4.2 Enrichment Factor

The enrichment factor (EF) was used to identify the potential sources, temporal variations, and accumulations of each element in the soil(Bian et al., 2013; Ministry of Environmental Protection et al., 2014; Tepanosyan et al., 2017). The EF is defined as the ratio of the elemental concentration to that in Earth's upper crust (Refence value). The calculation formula is given as follows:

$$EF = \frac{C_n/\mathrm{C_{ref}}}{B_n/B_{\mathrm{ref}}} \tag{2-2}$$

In this formula, C_n represents the concentration of the element of interest in soil, while C_{ref} is the concentration of the reference element in the same soil. B_n and B_{ref} represent the concentrations of the element of interest and the reference element in Earth's crust, respectively. The degrees of enrichment are classified into five categories: EF<2 represents minimal enrichment; 2≤EF<5 indicates moderate enrichment; 5≤EF<20 means significant enrichment, 20≤EF<40 indicates very high enrichment, while EF≥40 indicates extremely high enrichment. Generally, elements that are almost exclusively derived from the Earth's crust (Mn, Al, Fe, Sc, Ti, Zr, V, etc.) were chosen as the reference element(Anupam et al., 2016). The element, Mn, with a background concentration of 705 mg/kg in the soil in Beijing, was chosen as the reference element for this study(China National Environmental Monitoring Centre, 1990).

2.5 Health risk assessment

2.5.1 Hazard identification and exposure assessment

As shown in Fig. S1, CSM was developed based on a field survey conducted at residential playgrounds in Beijing. Several exposure pathways were considered according to the actual exposure scenarios, including oral ingestion, dermal contact with dust and soil, and inhalation through the respiratory system. It should be noted that Mn and Fe were not included in the assessment due to their high abundance in soil with negligible toxic effects to humans. The exposure model used in this study was preliminarily developed by U.S. EPA (Office of Emergency and Remedial Response Washington DC, 1989). The daily dose of soil or dust through oral ingestion, dermal absorption, and inhalation pathways was calculated by the following formulas. Exposure parameters were adapted from the U.S. EPA Exposure Factor Handbook(U.S. EPA, 1997).

Daily intake through oral ingestion:

$$OISER=\frac{OSIR\times ED\times EF\times ABS_o}{BW\times AT}\times 10^{-6} \tag{2-3}$$

where OISER represents the potential intake of soil or dust through oral ingestion, kg/(kg·d); OSIR is the daily intake of soil or dust, mg/d; ED is exposure duration, a; EF is exposure frequency, d/a; ABS_o is the oral absorption fraction, unitless; BW is body weight, kg; AT is the average time, d.

Daily intake through dermal absorption:

$$DCSER=\frac{SAE\times SSAR\times ED\times EF\times E_v\times ABS_d}{BW\times AT}\times 10^{-6} \tag{2-4}$$

$$SAE=239\times H^{0.417}\times BW^{0.517}\times SER \tag{2-5}$$

where DCSER is the intake of soil or dust through dermal absorption, kg/(kg·d);

SSAR is the adhesion coefficient of soil or dust to skin, mg/cm^2; The definitions of EF, ED, BW and AT are provided in formula (2-3); ABS_d is the dermal absorption factor, unitless; E_v is the daily dermal exposure frequency of an individual, time/d; SSAR is the skin adhesion factor, mg/cm^2; SAE is the surface area of body skin exposed, cm^2; SER is the ratio of exposed skin to the total skin area, unitless; H is the height of the individual, cm.

Daily intake through inhalation:

$$PISER = \frac{PM_{10} \times \mathrm{DAIR} \times ED \times \mathrm{PIAF} \times (f_{spo} \times EFO + f_{spi} \times EFI)}{BW \times AT} \times 10^{-6} \qquad (2\text{-}6)$$

where PISER is the intake of soil or dust through inhalation, kg/(kg·d); PM_{10} is inhalable particulate matter in ambient air, mg/m^3; DAIR is daily inhalation rate, m^3/d; PIAF is the retention ratio of inhaled particles; f_{spo} is the fraction of particles in outdoor air, unitless; EFO is the outdoor exposure frequency, d/a; f_{spi} is the fraction of particles in indoor air, unitless; EFI is the indoor exposure frequency, d/a; The definitions of ED, BW and AT are provided in formula (2-3).

2.5.2 Risk characterization

The non-carcinogenic risk through the three exposure pathways was characterized by the following formulas:

$$HQ_n = \sum_{i=1}^{n} HQ_i = HQ_{ois} + HQ_{dcs} + HQ_{pis} = \frac{OISER \times C_{sur}}{RfD_o \times SAF} + \frac{DCSER \times C_{\mathrm{sur}}}{RfD_d \times SAF} + \frac{PISER \times C_{\mathrm{sur}}}{RfD_i \times SAF} \qquad (2\text{-}7)$$

$$RfD_d = RfD_o \times ABS_{gi} \qquad (2\text{-}8)$$

$$RfD_i = \frac{RfC \times DAIR}{BW} \qquad (2\text{-}9)$$

where HQ_{ois} presents the non-carcinogenic hazard quotient of oral ingestion, unitless; RfD_o is the reference dose of oral ingestion, mg/(kg·d); HQ_{dcs} is the non-carcinogenic hazard quotient of dermal absorption, unitless; RfD_d is the reference dose of dermal absorption, mg/(kg·d); HQ_{pis} is non-carcinogenic hazard quotient for inhalation, unitless; RfD_i is the reference dose of inhalation, mg/(kg·d); RfC is the reference concentration, mg/m^3; DAIR is the daily inhalation rate, m^3/d; SAF is adherence factor, unitless; C_{sur} is HMs concentrations, mg/kg; ABS_{gi} is gastrointestinal absorption factor, unitless. The definitions of OISER, DCSER, PISER, C_{sur} and ABS_{gi} are provided in the formulas above.

The aggregated carcinogenic risks (CRs) through three exposure pathways were employed as following formulas:

$$CR_n = \sum_{i=1}^{n} CR_i = CR_{ois} + CR_{dcs} + CR_{pis} = OISER \times C_{sur} \times SF_o + DCSER \times C_{sur} \times SF_d + PISER \times C_{sur} \times SF_i \tag{2-10}$$

$$SF_d = \frac{SF_o}{ABS_{gi}} \tag{2-11}$$

$$SF_i = \frac{IUR \times BW}{DAIR} \tag{2-12}$$

where SF_o, SF_d and SF_i represent the slope factor of oral ingestion, dermal absorption and inhalation, repectively, mg/(kg·d); IUR is inhalation unit risk, m^3/mg. The definitions of OISER, DCSER, PISER, C_{sur} and ABS_{gi} are provided in above formulas.

In this study, the population was classified as adults and children based on their respective behavioral and physical characteristics. Land use was categorized as a residential area because the playgrounds were located within the residential communities. Toxic parameters for each element were collected from the U.S. EPA guideline on health risk assessment, as well as literature data. These parameters are moderately adapted according to the actual situation. Detailed parameters are summarized in Table S3 and Table S4.

2.5.3 Statistical analysis

Correlation analyses were conducted using Pearson's correlation analysis to illustrate the relationship between metal(loid)s in soil and dust and environmental variables, including the concentration of HMs, soil properties, and potential sources of contamination. Multiple statistical analyses, including correlation and descriptive statistics, were performed using the IBM SPSS software package, version 25.0, to identify potential emission sources affecting the health risk. The Upper Confidence Limit (UCL) was chosen to avoid overestimation of health risk results. Cumulative probability analysis and plots were generated using Origin 2021 software. Finally, the spatial distribution of health risk from exposure to HMs in residential playgrounds in Beijing was displayed using ArcGIS (version 10.0) software with kriging interpolation.

3 Results and discussion

3.1 The accumulation of heavy metal(loid)s in residential playgrounds

3.1.1 Geoaccumulation and enrichment

The results from Fig. 3, Table S5 and Table S6 indicated that the mean GIs of metal(loid)s in the urban soil were distributed in the following order: Cu>As >Ni>Mn>Co>Be>Cr>Se>Cd>Sb, whereas in suburban soil, the order was: As>Cu>Ni> Mn>Cr>Co>Be>Se>Cd>Sb. Despite the low GI values of almost all HMs (<0.36), the geoaccumulation of As and Cu in topsoil were relatively higher than that of the other metal(loid)s (Fig. 3a & b). Smith et al. suggested that the accumulation of As in urban soil was due to both lithogenic and anthropogenic sources(Smith et al., 1998). The mean levels of As in urban and suburban soil were 15.95 mg/kg and 15.26 mg/kg, respectively, several times higher than the As concentration in the Earth's crust (~1.5 mg/kg), providing the evidence of anthropogenic contributions to HMs accumulation in soil (Buttafuoco et al., 2016). As shown in Fig. 3a & b, the standard deviation of GIs of Cd in urban and suburban soil was 1.54 and 1.88, respectively, indicating high spatial variability among different residential communities. This variability might be attributed to fertilizer/pesticide application, irrigation with sewage water, and other activities involving Cd discharge(Kubier et al., 2019). The low background values of Sb (~1 mg/kg) likely accounted for the lowest GIs and EFs of Sb, as it typically bounded with oxides or hydroxides of Fe and Mn(Bolan et al., 2022).

Similarly, the high EFs of Ni, Cu, and As were observed in topsoil, while Be, Cr, Co, Se and Sb exhibited low EFs comparatively (Fig. 3c & d). The comparison of metal(loid)s EFs in Fig. 3c & d indicated a relatively higher enrichment of metal(loid) in suburban soil than in urban soil (Table S6). Previous studies suggested that industrial and agricultural wastes were the main anthropogenic sources of soil metal(loid)s(Xu et al., 2014; Yang et al., 2018). Thus, metal(loid)s accumulation might be caused by the collective effects of natural weathering of the Earth's crust, agricultural production, traffic, achitechive, municipal landfill, and industrial wastes, etc.(Alloway 2013; Atafar et al., 2010; Bradl 2005; Shen et al., 2018). In particular, intensive industrial and agricultural activities in suburban areas might contribute to the high accumulation of trace elements in soil to some extent.

(Fig. 3)

3.1.2 Comparison with previous studies

An analysis of the similarities and differences in metal(loid)s in relation with different environmental media, land use types and locations provided a preliminary understanding of the overall geoaccumulation status and underlying influencing factors (Table 1). The higher concentrations of metal(loid)s in dust, compared to those in soil in same city, reflected the stronger adsorption capacity of dust particles towards HMs(Javid et al., 2021). Atmospheric dust was known for prominent adsorption abilities due to its large surface areas(Li et al., 2013), porous structure(Hu et al., 2012), organic materials and oxygen-containing functional groups(Hamanaka et al., 2018), which enable it to retain HMs pollutants. As shown in Table 1, present study demonstrated that metal(loid)s concentration in soil was below the risk screening values. However, the concentrations of Ni, Cu and As all exceeded corresponding background values by different degrees. This finding was consistent with similar studies where significant metal(loid)s were detected in urban soil/dust, but only a few metal(loid)s posed a slightly unacceptable risk(Pan et al., 2018).

(Table 1)

3.2 Source identification of heavy metal(loid)s in residential playgrounds

3.2.1 Correlation of heavy metal(loid)s concentrations with environmental variables

Environmental variables, such as land use, vegetation coverage, bare soil areas, and distance to main road, construction site, factories, highway, were recorded for assessing their correlation with metal(loid)s concentrations (Table S7). Pearson's correlation analysis revealed a weak positive correlation between soil/dust metal(loid)s levels with environmental variables (maximum correlation coefficient of 0.435, *p value* <0.01). Specifically, weak positive correlations at the 0.01 level were found between the distance to highways and the levels of Be, Cr, dust Be in soil, while negative correlations at the 0.05 level were observed between bare soil area with the levels of Cr, Mn, Cu in dust. These positive correlations suggested that airborne particulate matter (PM) had a significant effect on the accumulation of metal(loid)s in equipment dust. For example, HM emissions from traffic vehicles may deposit on the surface of playground equipment after long-distance diffusion and transport(Chen et al., 2010). The observed significant negative correlation between the extent of bare soil areas and the concentration of metal(loid)s in dust may be attributed to the vegetation's ability to retain atmospheric particulate matter (PM). Specifically, playgrounds with lower vegetation density are associated with weaker interception capability, resulting in increased PM deposition on recreational equipment(Liu et al., 2018).

3.2.2 Correlation between metal(loid)s concentration in soil and dust

Frequency distribution and Pearson's correlation analysis (with a *P value* <0.01) were performed to illustrate the As/Ni concentration and their correlation with Mn/Fe in the same environmental media (Fig. 4, Fig. S4 & Fig. S5). Fig. 4a & d displayed the relative frequency of different ranges of As/Ni concentration in soil, while Fig. 4b, c, e and f focused on the significance of their relation with Mn/Fe in soil.

The results revealed a considerable contrast in both soil and dust. In soil scenarios, a significant positive correlation was observed between the concentration of As and Mn/Fe, with the coefficients of 0.358 and 0.265, respectively, while were higher than those in dust (only 0.185 and 0.042). Similarly, the correlation coefficients between concentration of Ni to Mn/Fe in soil reached 0.463 and 0.575, respectively, which were higher than those in dust (only 0.097 and 0.033). Pearson's correlation indicated As/Ni in soil or dust was mostly absorbed by Mn and Fe oxides, which was consistent with previous studies(Alves et al., 2011; Cai et al., 2002; Mahimairaja et al., 2005; Massoura et al., 2006; Yang et al., 2019). No significant correlation was observed in levels of HMs in soil and dust(Fig. S3), indicating that the HMs in dust might be contributed by multiple types of pollution sources other than local soil contamination.

(Fig. 4)

3.3 Health risk of metal(loid)s exposure at residential playgrounds

3.3.1 Cumulative probability of health risk

In this study, the assessment of cancer risk resulting from single metal(loid) exposure and combined CRs from all trace metal(loid)s was conducted using USEPA dose-response models and a site-specific CSM (section 2.5 and Fig. S1). Carcinogenic metal(loid)s in dust associated with epigenetic carcinogenesis in DNA disruption (such as As, Cd and Ni) were considered in the assessment of carcinogenic risk. The CRs for a single exposure pathway and the combined CR from exposure to As, Cd, and Ni in dust were presented in Table S8. Fig. 5 illustrates the carcinogenic health risk of exposure to heavy metal(loid)s in dust as cumulative probability. The acceptable level was represented as a dotted line in subfigures. The estimated results showed a range of combined risk of 6.87×10^{-7} to 6.22×10^{-6}, and 1.03×10^{-6} to 8.36×10^{-6} to children, and adults, respectively. Almost all of the dust metal(loid)s except Cd had potential carcinogenic effects on local residents, with the mean risk of 2.47×10^{-6} and 3.49×10^{-6} to children and adults, respectively. These values all exceeded the U.S. EPA's safe limit of 1.00×10^{-6}. The predominant exposure pathways for these CR appeared to be ingestion and inhalation(Iwegbue et al., 2018; Sabouhi et al., 2020). Ingestion and inhalation contributed approximately 77.73% and 16.56% of children's CR, respectively, while for adults, these values were 68.19% and 28.17%, respectively.

(Fig. 5)

A similar cumulative probability profile was observed in the CR of soil metal(loid)s exposure, with combined CRs up to 5.15×10^{-6} and 7.23×10^{-6} for children and adults, respectively (Fig. 6), and mean combined CRs of 3.75×10^{-6} and 5.29×10^{-6}, respectively (Table S10). A potential cancer risk to residents was indicated after their lifetime exposure to HMs in soil. Almost the entire combined CR was contributed by exposure to As and Ni, making them high-priority metal(loid)s requiring control measures to address the carcinogenic effects. This was despite the fact that total As concentration in soil in study area was much lower than those previously reported in the literatures for other types of lands. For example, the average concentrations of total As in soil around mining area, coking area, metal smelting and chemical plants were all above 51.51 mg/kg. However, the mean soil As level in present study was only 15.56 mg/kg, which still exceeded the safety threshold value (8.0 mg/kg in soil) recommended by the World Health Organization(Liu et al., 2016). This again validated the possible health risk caused by long-term exposure to As in soil in residential playgrounds, emphasizing the need for effective control.

(Fig. 6)

The method introduced in Section 2.5 was used to quantify the non-carcinogenic effects of exposure to HMs at a playground scenario. HQ, the ratio of metal(loid) exposure and the reference dose (RfD), was initially obtained by toxicological experiments of metal dose-response experiments in animals. As shown in Fig. S6 and Fig. S7, most of the HQs of single HM exposure at residential playgrounds were below the acceptable HQ limit recommended by the U.S. EPA (HQ=1), but the limit was exceeded at a few sites. This suggested that the non-carcinogenic effects on residents' health might be negligible when considering single metal(loid)s exposure. Unfortunately, when considering all of the metal(loid)s, the maximum HQs of dust and soil metal(loid)s exposure reached 1.98 and 1.44, respectively, exceeding the safety risk limit. Because children were vulnerable and sensitive to environmental toxicants, higher HQs of children than adults were observed in Fig. S6 and Fig. S7. As shown in Table S9 and Table S11, the combined HQs of children's dust metal(loid) exposure were distributed in the following order: Se<Cu<Be<Sb<Cd<Ni<As<Cr<Co, while that in soil exposure scenarios were: Se<Sb<Cu<Cd<Be<Ni<Cr<As<Co. This slight variation was likely due to the cumulative toxicities associated with the enrichment of multiple metal(loid)s in dust/soil. The overall risk assessment results showed that children, as receptors relatively more sensitive to non-carcinogenic risk, were less resistant than adults to the non-carcinogenic effects of HMs exposure at residential playgrounds. The final risk was influenced by a range of factors, such as age groups, behavior patterns, exposure durations, diverse residential communities, and individual personalities of children. Thus, it was crucial for future studies to account for these variable factors in environmental risk assessment.

3.3.2 Spatial distribution of health risk

The spatial distribution of CRs and HQs of children's metal(loid)s exposure at residential playgrounds was analyzed by kriging interpolation with ArcGIS (Fig. 7). As shown in Fig. 7a and b, it was observed that the sites exhibiting high CR and HQ values of HMs exposure in soil were predominantly clustered in the areas bordering the urban periphery, specifically in the vicinity of the 5th Ring Road of Beijing, and in suburban counties in the northeast region of this city. The spatial distribution patterns of health risk were highly correlated with the concentrations of HMs in soil. The high risk concentrated in the urban boundary was mostly attributed to heavy traffic, resulting in significant vehicular emissions of HMs. The distinctive topography of Beijing might also contribute to the health risk of soil HM exposure. Beijing is encircled by several mountains on its north, west, and east sides, while the plains of North China are only present in the south. On these three sides, the city is bounded by the Taihang Mountains and Yan Mountains. HMs in the atmosphere, carried by winds from the south, can deposit onto the ground in the northern, eastern, and western areas of the urban region, accumulating in the soil near the city boundaries. High health risk in suburban areas was ascribed to high metal contents in soil parent materials and agricultural production activities, whereas that in urban area was probably related to the anthropogenic sources including traffic emission, atmospheric deposition, industrial exhaustive gas, etc. (Table S7)(Fan et al., 2021; Wei et al., 2010). The spatial distribution of health risk associated with dust exposure are presented using spatial interpolationg, as well. As shown in Fig. 7c and d, a little difference between the risk by soil and dust exposure were that the high health risk of dust HMs exposure was mainly distributed in the northwest of suburban area and eastern boundary of urban area. The phenomenon might be attributed to the complex origins of HMs in equipment dust and soil. Traffic emissions, soil dust, industrial emissions, fossil fuel combustion, natural wind, and dust storms all served as significant sources of HMs in equipment dust, and meteorological conditions further influenced the migration and diffusion of HMs gaseous pollutants(Soleimani et al. 2018). The high risk concentrated in the urban boundary was mostly attributed to heavy traffic, resulting in significant vehicular emissions of HMs, while the high dust heavy metal risk in the northwest region may be associated with industrial sources and wind convection in the southeast direction. Relevant meteorological records demonstrated that Beijing frequently experienced strong southeast winds during summers(Tie et al. 2015). Therefore, the high soil HM risk in the northwest region (Fig. 7d) might be associated with industrial sources and wind convection in the southeast direction.

(Fig. 7)

3.4 Relationship between health risk and comprehensive parameters

The correlation between health risk and relevant parameters at residential playgrounds, including environmental, economic and social aspects, was analyzed using Pearson's Correlation. A series of statistical data, including population, pollutant concentration, vehicle ownership, GDP, etc., was collected from the Beijing Yearbook 2021. The correlation between health risk and comprehensive statistical data could provide appropriate basis for policy making associated with risk migration at residential playgrounds.

The analytical results and original data are presented in Fig. 8 and Table S12, respectively. Economic factors might indirectly influence the health risk at these sites, as they affected factors such as urbanization, industrialization, and transportation, which were known to contribute to heavy metal pollution. A positive correlation was observed between pollutant concentration and the socio-environmental parameters, such as vehicle ownership, energy consumption, completed construction area, and GDP. It was evident that rapid economic growth resulting from energy consumption had led to increased vehicle ownership, and subsequently, the emission of high concentrations of SO_2, NO_2, PM, etc., into the air around roads or highways with high traffic volumes from vehicle exhaust. Interestingly, a significant negative correlation between soil HQs (S-HQ) and air pollutant concentration or construction areas suggested a negligible relationship between the accumulation of soil metal(loid)s and deposition of air pollutants. Moreover, it might have an intensive relation with the areas under construction rather than the completed built-up area. Another noteworthy result was that the risk of soil metal(loid)s was positively correlated with radius, indicating that the higher the health risk from soil metal(loid)s, the further away from urban centers. It was also explicitly verified by the spatial distribution patterns in Fig. 7a and b, where the high soil risk was concentrated around 5th Ring Road but low soil risk was located in the city center. Anthropogenic activities, such as dense construction, transport volumes, and small amounts of agricultural production located in suburban areas, contributed to the metal(loid)s accumulation in soil with different degrees. The high concentrations of metal(loid)s in suburban soil were likely due to the urban heat island effect combined with the blockage of natural ventilation by high-risk buildings in urban areas(Wang et al., 2021).

(Fig. 8)

3.5 Environmental implications for risk management

In the present study, the chronic exposure of children or adults to HMs accumulated in soil or equipment dust at residential playgrounds may be frequently overlooked. Therefore, greater attention should be paid to accurate risk quantification followed by precise assurance of health safety of residential playgrounds where the HMs exposure is of high frequency for local residents, especially for more sensitive children. To accurately assess health risk, it is also necessary to clarify the accumulation of metal(loid)s in the dust or soil of residential playgrounds. On-site detection technologies can be integrated with risk management to enhance the efficiency of on-site environmental quality monitoring and decision-making effectiveness. Several technologies such as electrochemical detection systems, laser-induced breakdown spectroscopy, X-ray fluorescence can be employed for swift on-site monitoring of soil quality in playgrounds, thereby providing a foundation for risk prediction and control decision-making. Then the correlation of risk with environmental variables can serve as the foundation for further source apportionment and risk control. In present study, significant accumulation of As and Ni in soil and equipment dust was observed, making them the metal(loid)s with the highest priority for control in the residential playground environment. Therefore, some recreational playgrounds in Beijing may have unacceptable carcinogenic effects on residents. To address this potential risk, efforts are required to identify the underlying effects and implement suitable measures for risk control. Among all of the measures, soil quality standards provide acceptable threshold values of each metal(loid), upon which preliminary contamination evaluations are conducted to determine control priority. Many countries, including China, still lack soil risk management standards, especially at specific scenarios of residential playgrounds. Thus, it is imperative to conducton-site investigations and establish standards for HMs, especially the metal(loid)s with high-priority, to facilitate the construction of standard systems. However, a serious challenge remains regarding the variation of quality standards for soil metal(loid)s, which are the combined effects of multiple factors throughout the development processes, including land-use type, soil properties, cancer endpoint, and protection targets (i.e. sensitive risk receptors). Therefore, it is necessary to conduct more comprehensive and standardized risk assessments that take into account many relevant factors, and to establish more specific and tailored standards for different land-use types and sensitive risk receptors.

4 Conclusions

This study provides valuable insights into the health risk of HMs exposure at residential playgrounds in mega-cities. Results indicated that significant accumulation of As, Cu, and Ni in soil, especially in suburban residential communities. Although, the mean concentrations of As, Ni, Cu, and Cd in soil were less than risk screening values, they were higher than corresponding background values. Correlation analysis revealed that As/Ni in soil significantly bound with Mn/Fe oxides in parent materials, while the accumulation of other metal(loid)s in soil and dust could be ascribed to the cumulative impacts of traffic emission, deposition of atmospheric particulate matter, and other sources. A site-specific CSM and assessment model was developed for quantitatively characterizing health risk of heavy metal exposure at residential playgrounds. The assessment results indicated the mean CRs of dust metal(loid)s exposure were 2.47×10^{-6} and 3.49×10^{-6} to children and adult, respectively, and 3.75×10^{-6} and 5.29×10^{-6} at soil exposure scenarios, respectively. The slightly unacceptable CRs to children were primarily ascribed to the high levels of lithogenic As and Ni, while the negligible unacceptable total HQs was due to exposure to As, Co, Cr. Spatial interpolation revealed that health risk increased with distance to the city center, suggesting that high risk mainly concentrated in suburb rather than urban areas. This study emphasizes the need for effective decision-making and risk control, particularly for As, Ni, and other metal(loid)s of priority concern, to ensure a safer and cleaner recreational environment for residents. Thus, an intensive attention is required for sound decisions with regard to effective risk control of HMs exposure at a playground scenario. The findings can be regarded as a valuable reference for urban environmental management, despite the acknowledgment of some limitations, such as the limited analysis of metal(loid)s and the narrow focus on health risk associated with metal(loid)s exposure alone, without considering other pollutants.

Acknowledgements

This work was supported by GDAS' Project of Science and Technology Development (2022GDASZH-2022010105), National Natural Science Foundation of China (42177195), National Key R&D Program of China (2020YFC1806301-2), the Science and Technology Foundation of Guangzhou (202201011514), Guangdong Basic and Applied Basic Research Foundation (2022A1515110625), and China Postdoctoral Science Foundation (2022M720848). We sincerely acknowledge the Center for Environmental Quality Test at Tsinghua University for their valuable support in the analysis of metal(loid)s. We are grateful for the academic exchanges with colleagues who have inspired the authors with critical ideas presented in this work. We greatly appreciate the technical assistance provided by Assistant Prof. Shovan Kumar Sahub. The authors also extend their gratitude to the anonymous reviewers for their critical review and constructive suggestions for this work.

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Table caption

Table 1 Comparison with the concentration of heavy metal(loid)s in (dust) in other cities (mg/kg)

Table 1. Comparison with the concentration of heavy metal(loid)s in soil (dust) in other cities (mg/kg)

Location	Land types	Media	Year	Cr	As	Ni	Cu	Cd	Zn	Ref.
Yanta District, Xi'an City, China	Leisure Squares	Dust	2016	395.57	NA	42.74	85.61	8.68	NA	(Shao et al., 2018)
Jiaozuo City, Henan province, China	School and urban park	Soil	2016	58.53	57.68	35.79	20.88	0.53	99.51	(Han et al., 2020)
Jiaozuo City, Henan province, China	School and urban park	Dust	2016	112.07	23.08	51.07	49.85	1.25	374.30	(Han et al., 2020)
Dhaka, Bangladesh	City street	Dust	2017	144.34	8.09	37.01	49.68	11.64	239.16	(Safiur Rahman et al., 2019)
Yerevan, Armenia	Urban area	Soil	2012	122.00	0.78	61.90	103.00	NA	262.00	(Tepanosyan et al., 2017)
Ranga Reddy Province and Telangana State, South India	Urban region	Soil	2019	43.24	NA	7.55	40.64	NA	34.68	(Adimalla et al., 2020)
Villavicencio, eastern Colombia	Highway	Dust	2014	9.40	NA	5.30	126.30	NA	133.30	(Trujillo-González et al., 2016)
Villavicencio, eastern Colombia	Residential community	Dust	2014	7.30	NA	7.20	23.70	NA	108.30	(Trujillo-González et al., 2016)
Villavicencio, eastern Colombia	Residential community	Dust	2014	60.20	NA	54.30	490.20	NA	387.60	(Trujillo-González et al., 2016)
Villavicencio, eastern Colombia	Background	Dust	2014	40.20	NA	13.20	35.10	NA	59.90	(Trujillo-González et al., 2016)
Thessaloniki, Greece	Congested area	Dust	2009	104.90	NA	89.43	662.30	1.76	452.80	(Bourliva et al., 2017)
Qingdao City, China	Urban Street	Dust	2017	153.10	NA	60.90	456.70	0.75	708.30	(Xu et al., 2019)
Beijing City, China	Residential playground	Soil	2017	**54.70**	**15.56**	**33.72**	**43.42**	**0.07**	**NA**	Present study
Beijing City, China	Residential playground	Dust	2017	**78.88**	**10.02**	**22.26**	**52.07**	**0.72**	**NA**	Present study
Background values, Beijing	All types	Soil	1990	68.10	9.70	29.00	23.60	0.07	NA	(China National Environmental Monitoring Centre 1990)
Risk screening values	Residential land	Soil	2018	NA	20.00	150.00	2000.00	20.00	NA	(Ministry of Ecology and Environment of the People's Republic of China 2018)

Figure Click here to access/download;Figure;Figure list.docx

Figure captions

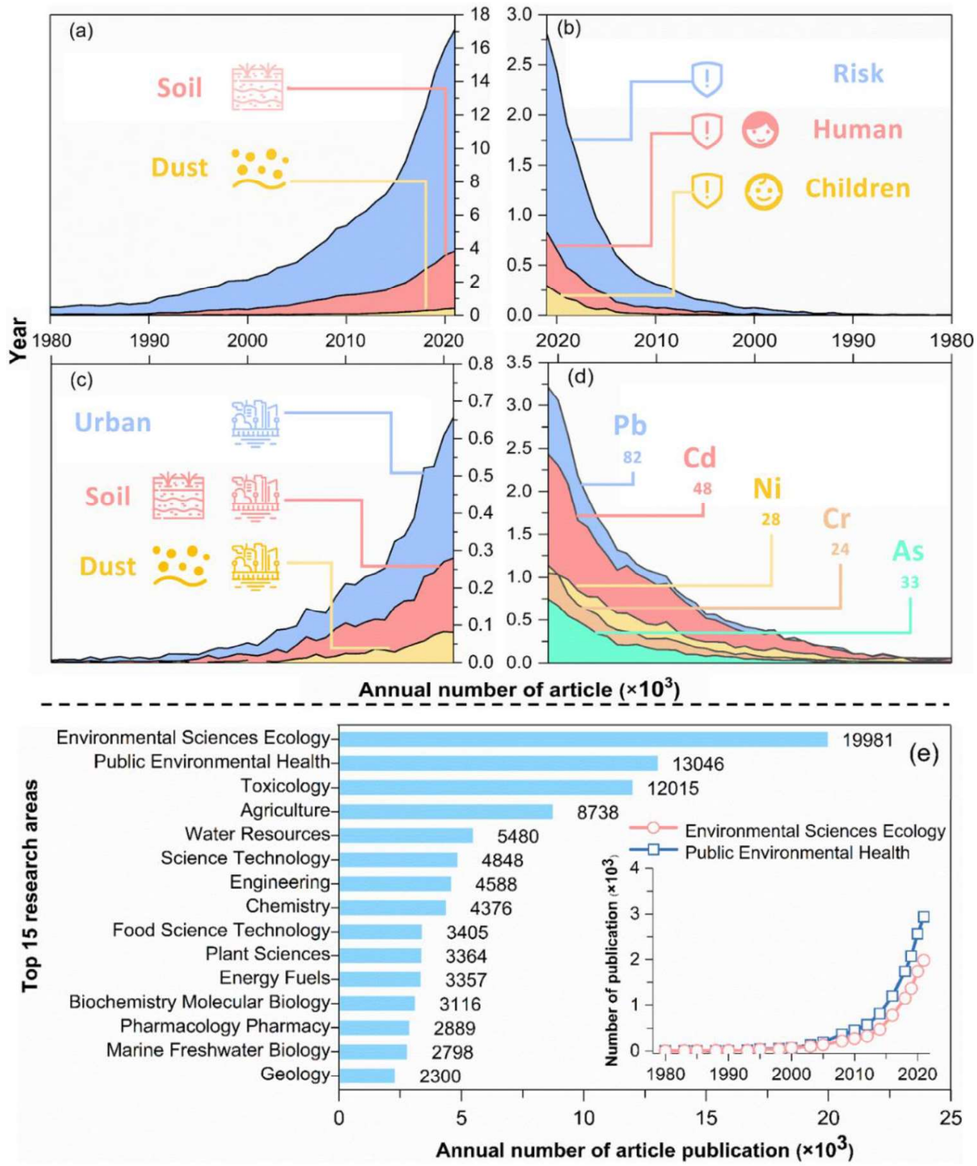

Fig. 1. A 40-year review of research on the environmental risk associated with heavy metals

(The trend of the numbers of published literature derived from a keyword search in Web of Science Database, including a, “heavy metal”, “heavy metal” and “soil/dust”; b, “heavy metal” and “risk”, “heavy metal” and “risk” and “human/children”; c, “heavy metal” and “urban”, “heavy metal” and “urban” and “soil/dust”; d, “heavy metal” and “lead/cadmium/nickel/chromium/arsenic”; the search was conducted in September 2022)

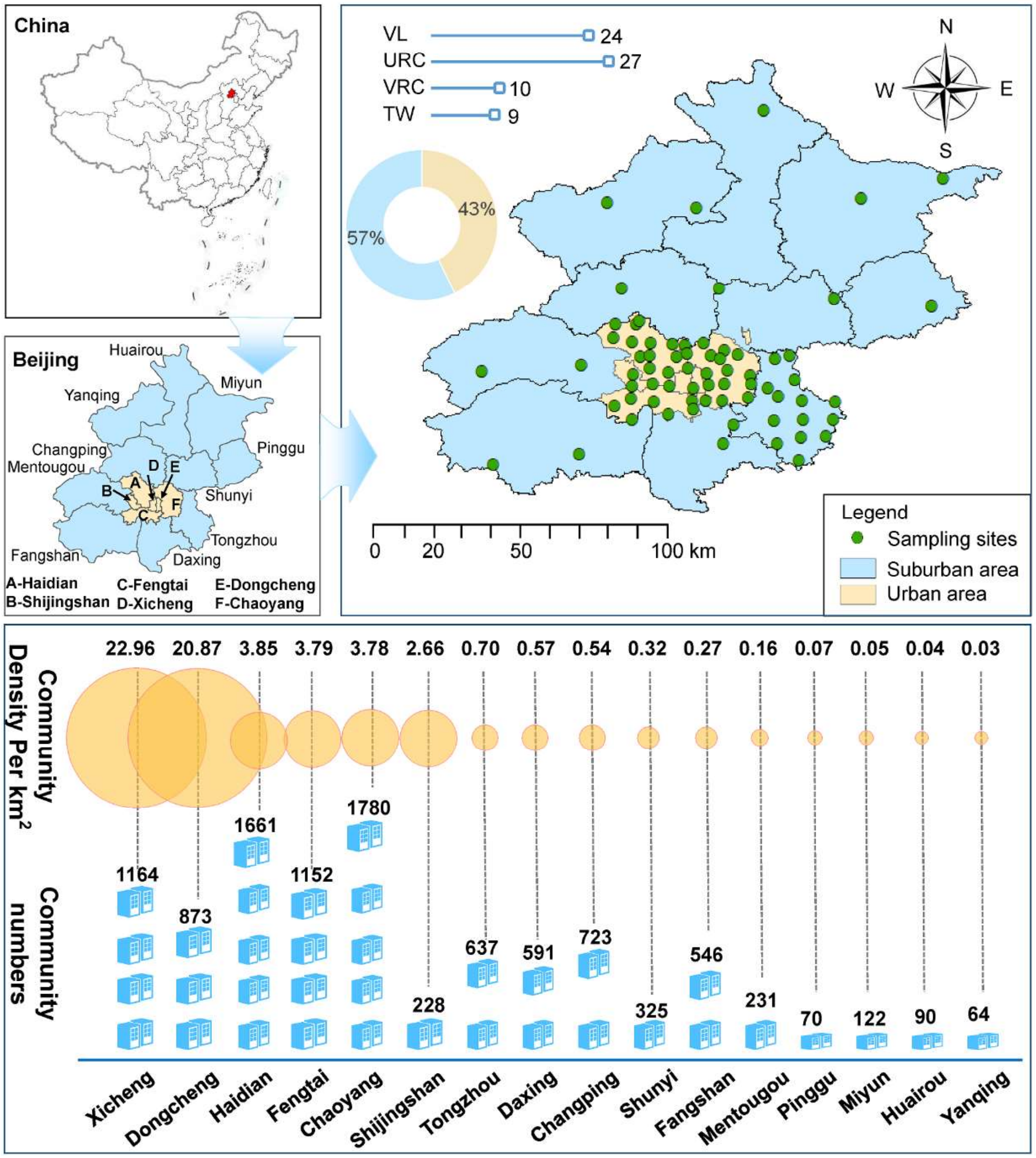

Fig. 2. Distribution of sampling sites and residential communities across the study areas

(VL, Village Land; URC, Urban Residential Community; VRC, Village Residential Community; TW, Town; The map was creased using ArcGIS 10.0 ESRI; The total area of each district was obtained from the Beijing Municipality Government, and the number of communities was sourced from a housing service platform called Beijing Beike Technology Co., Ltd.)

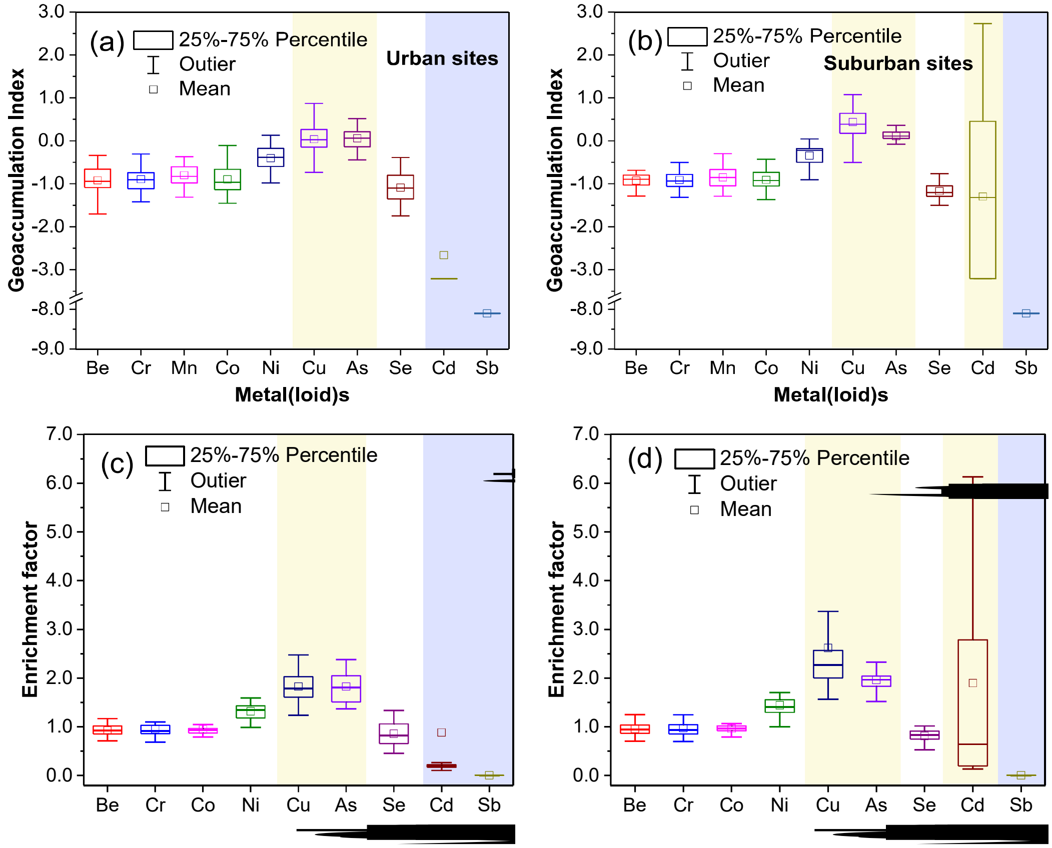

Fig. 3. Box-whisker plot for GIs and EFs of metal(loid)s in soil

(a & c, urban sites; b & d, suburban sites; GIs, Geoaccumulation Indexes; EFs, Enrichment Factors)

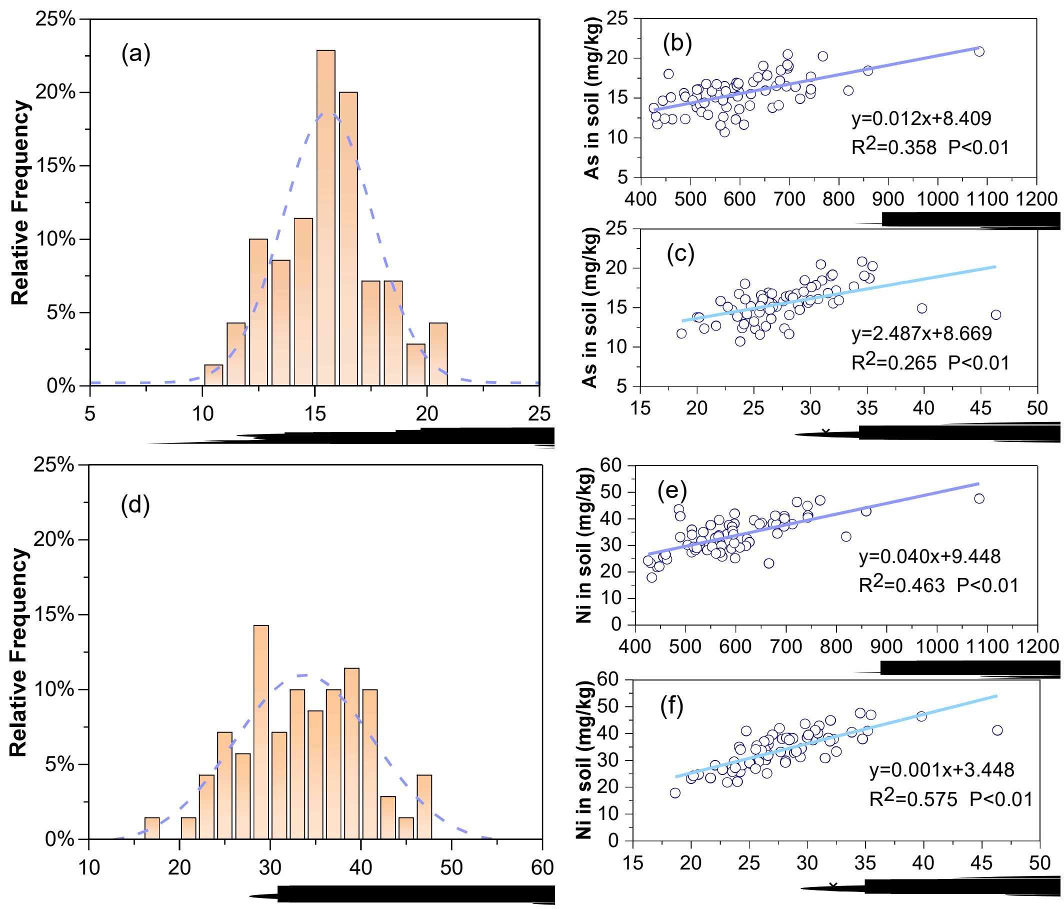

Fig. 4. Distribution of arsenic and nickel concentration in soil of the study area

(a, histogram of soil arsenic concentration; b & c, correlation between the concentration of As and the concentration of Mn/Fe in soil; d, histogram of soil nickel concentration; e & f, correlation between the concentration of nickel and the concentration of Mn/Fe in soil)

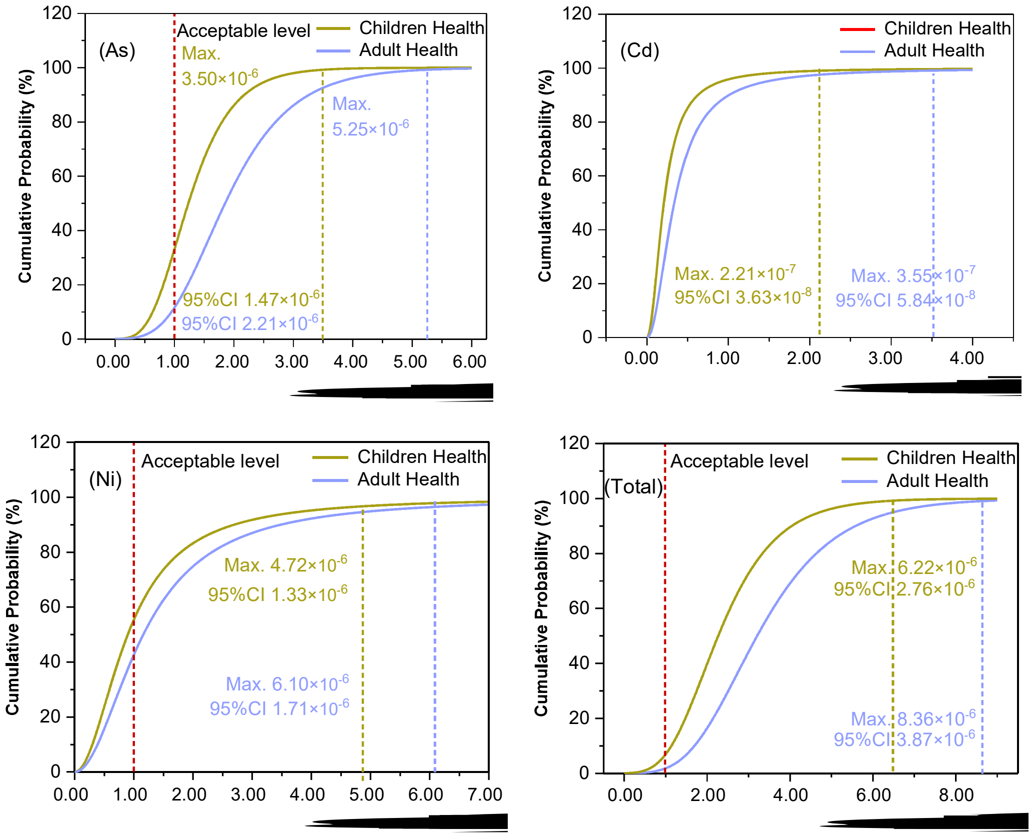

Fig. 5. Cumulative probability of carcinogenic health risks of exposure to heavy metal(loid)s in dust at residential playgrounds

(Receptors include children and adult; 95%CI, 95% upper limit of confidence interval)

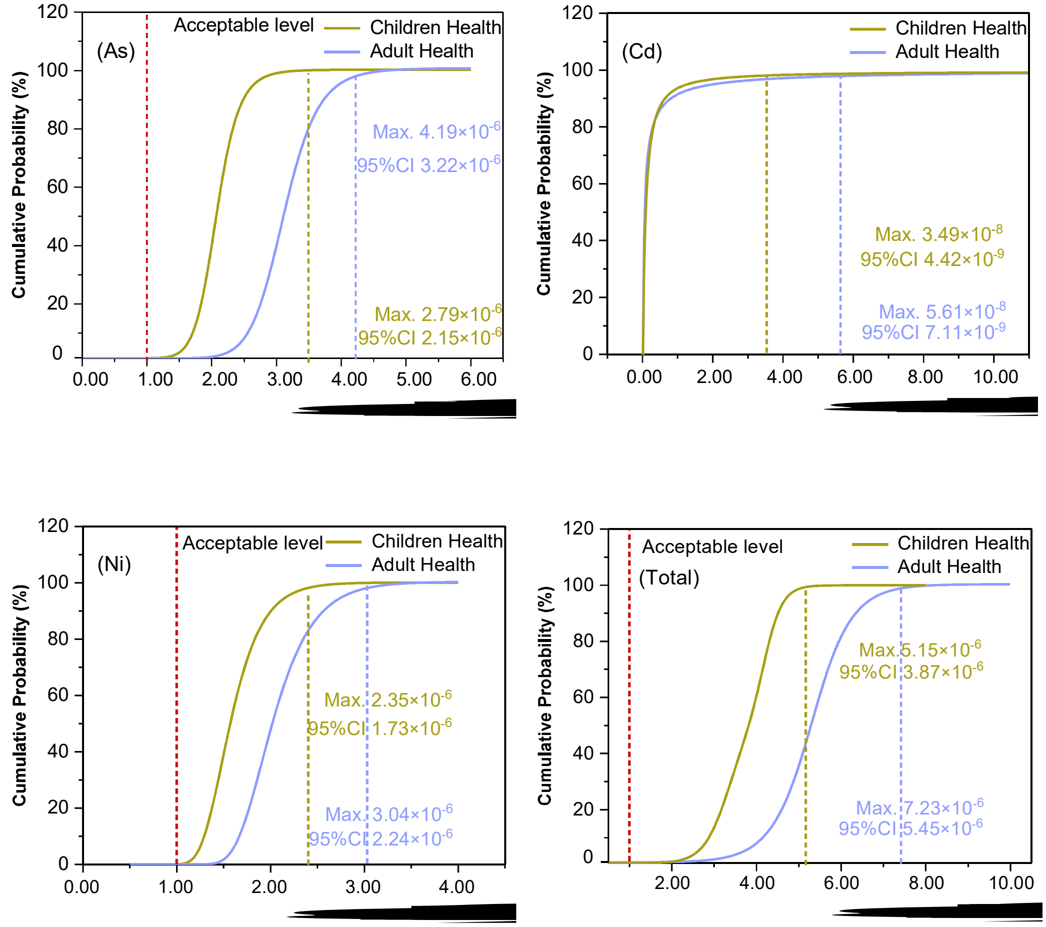

Fig. 6. Cumulative probability of carcinogenic risks of exposure to heavy metal(loid)s in soil at residential playgrounds

(Receptors include children and adult; 95%CI, 95% upper limit of confidence interval)

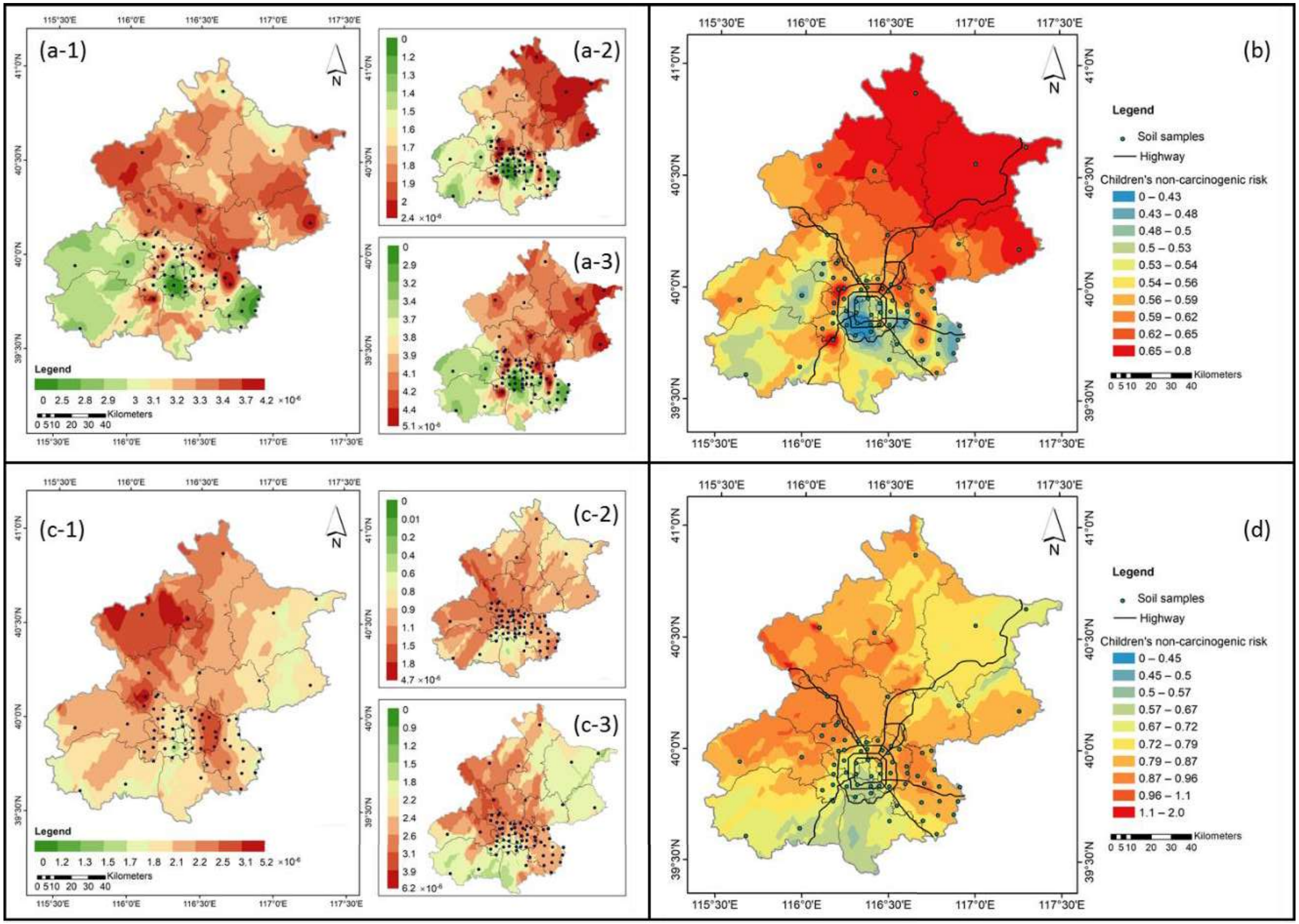

Fig. 7. Spatial distribution of CRs and HQs of heavy metal(loid)s exposure for children at residential playgrounds in Beijing

(a-1, comprehensive CRs by playground soil; a-2, CRs by As in playground soil; a-3, CRs by Ni in playground soil; b, comprehensive HQs by playground soil; c-1, comprehensive CRs by playground dust; c-2, CRs by As in playground dust; c-3, CRs by Ni in playground dust; d, comprehensive HQ by playground dust)

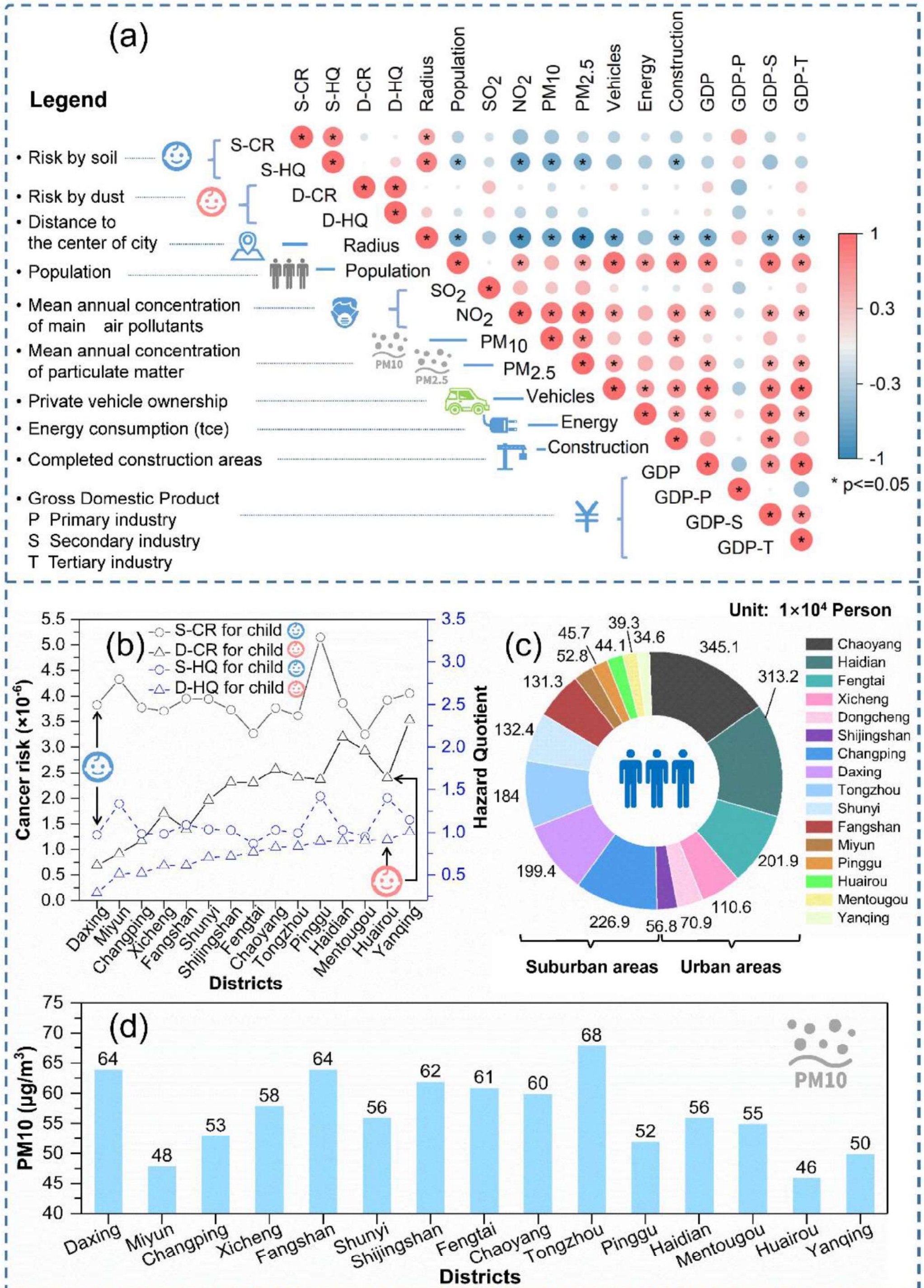

Fig. 8. Correlation between children's health risk and statistical indicators

(a, correlation between children's health risk with statistical indicators; b, health risk for children exposed to heavy metal(loid)s in soil and dust, including carcinogenic risks (CRs) and non-carcinogenic hazard quotients (HQs); c, distribution of the population across different administrative districts within Beijing; d, annual mean concentration of particulate matter in different administrative districts of Beijing; All of the statistical data used for correlation analysis is from the Beijing Statistical Yearbook 2021. S-CR refers to the carcinogenic risk from soil; S-HQ means the non-carcinogenic hazard quotient from soil; D-CR refers to the carcinogenic risk from dust; D-HQ means the non-carcinogenic hazard quotient from dust; “tce” denotes the unit of energy consumption and represents “ton of coal equivalent”)