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# Significant urban hotspots of atmospheric trace element deposition and potential effects on urban soil pollution in China

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ABSTRACT

Rapid urbanization has profoundly altered the spatial patterns of multiple element cycles. Whether and how urbanization shapes the spatial patterns of atmospheric trace element deposition remains, however, poorly understood. Using a newly compiled database on bulk deposition of eight trace elements (i.e., Cu, Ni, Zn, As, Cd, Cr, Hg and Pb) in China, we assessed the urban imprints on the spatial patterns of trace element deposition. Bulk deposition of the eight trace elements all showed a significant increase with closer distance to the nearest large cities, while the urban effect was also mediated by point emission sources and precipitation. We further compiled a database of urban topsoil (0–10 cm) concentrations of the eight trace elements and found that urban soil quality standards were exceeded in 80% of the studied cities for Cr, 49% for As, and less than 25% for other trace elements, respectively. The urban topsoil concentrations of six trace elements (except As and Hg) showed no significant correlations with their background values for natural soils, while we found a significant correlation between bulk deposition and urban topsoil concentrations of trace elements corrected by background values. We also demonstrated that current levels of trace element deposition would substantially increase urban soil pollution over the coming decades. Our findings confirm the occurrence of urban hotspots of trace element deposition and their impact on soil pollution and highlight a need of emission control of trace elements for safety urban soil quality.

## 1. Introduction

Unprecedented urbanization has occurred over the globe and profoundly altered biogeochemical cycles of various elements (Sen and Peucker-Ehrenbrink, 2012; Sun et al., 2020; Liu et al., 2020). Urban areas are hotspots of anthropogenic emissions due to intensive energy production, motor traffic, waste treatment, construction and industrial activities (An et al., 2019; Liu et al., 2021; Sun et al., 2017). Consequently, atmospheric deposition of many macro-elements, such as nitrogen (N), phosphorus (P), sulfur (S), potassium (K), calcium (Ca) and magnesium (Mg), are found to increase with closer distance to large cities (Bettez and Groffman, 2013; Du et al., 2015, 2016, 2018; Decina et al., 2020), affecting urban soil and water quality via acidification, nutrient imbalances and eutrophication (Hobbie et al., 2017; Du et al., 2022). Atmospheric deposition of trace elements, including micronutrients (e.g., Cu, Ni, Zn) and toxic heavy metals (e.g., As, Cd, Cr, Hg, Pb), can also have important impacts on environment (Zhong et al., 2020) and/or human health (Cao et al., 2012). Although the global utilization of various trace elements has increased rapidly for centuries (Penuelas et al., 2022), knowledge gaps remain in whether and how urbanization alters the spatial patterns of atmospheric deposition of trace elements.

Trace elements in atmospheric deposition are derived from both natural sources and anthropogenic processes. Urbanization and industrialization can lead to increased trace element emission and deposition in urban areas (Sen and Peucker-Ehrenbrink, 2012; Pecina et al., 2020). Major anthropogenic sources of trace metals in urban areas include construction and road wear, vehicle mechanical abrasion of brakes and tires, exhaust emissions, widespread use of fossil fuels, and the combustion of municipal solid waste (Klimont et al., 2017; Yun et al., 2020;

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Fussell et al., 2022). Atmospheric deposition fluxes of multiple trace elements are thus expected to increase with closer distance to the urban areas but the hypothesized urban hotpots of trace element deposition remain unexamined.

The urbanization induced spatial pattern of trace elements deposition can be potentially mediated by point emission sources outside central urban areas as well as climate factors. For instance, industrial activities, mineral extraction, coal combustion, and agricultural tillage practices, which are not usually distributed in central urban areas, are important point sources of trace elements (Hu and Cheng, 2013). Moreover, precipitation is an important climatic factor to promote atmospheric deposition of trace elements and reduce their transport distance (Pan and Wang, 2015). However, it remains unknown how these factors mediate the hypothesized urban hotspots of trace element deposition.

Atmospheric deposition may contribute to trace element accumulation in surface soils and cause various negative effects on soil (e.g., disturbing the balance of important soil processes) (Amundson et al., 2015), plant (e.g., inhibiting photosynthesis and growth) (Andresen et al., 2018), food safety (Hou et al., 2020), and human health (e.g., damage to organ function, immune, nervous, and endocrine systems) (Khanam et al., 2020; Zhang et al., 2021). Therefore, a number of countries have promulgated safety standards to identify pollution risks caused by soil trace element (MEEC, 2018a; MEEC, 2018b; VROM, 2000; CCME, 1999; US EPA, 1994; ICRCL, 1987; Ministenum et al., 1991; Eikmann, 1991; CCME, 1991; De Vries et al., 1993). Particularly, urban surface soils are an important sink of atmospheric deposited trace elements(Antoniadis et al., 2017; Yuan et al., 2021). However, the current status of trace element pollution for urban soils and potential risks due to high-level atmospheric deposition remains poorly understood in large areas of the world.

China has experienced very rapid urbanization and economic development in last several decades (Zhang et al., 2022) accompanied with a strong increase in anthropogenic emissions of trace elements (Tian et al., 2015). By compiling a national database on bulk deposition of eight trace elements (i.e., Cu, Ni, Zn, As, Cd, Cr, Hg and Pb) from published literature, we tested the hypothesis of urban hotspots of trace element deposition and explored their potential mediators. Additionally, we established a national database of urban topsoil (0–10 cm) concentrations of the eight trace elements in large cities (urban population $>$ 0.5 million). By comparing urban soil concentrations of trace elements with urban soil quality standards, we assessed the current status of urban soil trace element pollution. To evaluate the potential impact of trace element deposition on urban soil pollution, we further tested the correlations between urban topsoil concentrations of trace elements and corresponding bulk deposition to urban areas. To assess the potential confounding impact of variations in soil background values, we also tested the relationship between current urban topsoil concentrations of trace elements and their corresponding background values for natural soils. Finally, we also conducted a scenario analysis to demonstrate the potential contribution of bulk deposition to urban soil trace element pollution.

## 2. Materials and methods

### 2.1. Datasets

#### 2.1.1. Bulk deposition of trace elements

We compiled a national database of bulk deposition of eight trace elements (e.g., Cu, Ni, Zn, As, Cd, Cr, Hg and Pb) in China by searching literature via the Web of Science (http://isiknowledge.com) and the China National Knowledge Infrastructure (http://www.cnki.net). The keywords 'trace element'/'heavy metal', 'bulk deposition'/'atmospheric deposition' and 'China' were used. Target data were either directly taken from tables or digitized precisely from figures/plots using a GetData Graph Digitizer (Version 2.25, http://www.getdata-graph-digitizer.com). Bulk precipitation samples were commonly collected using continuously open samplers and bulk deposition was calculated based on annual bulk precipitation and precipitation volume-weighted mean trace element concentrations of the bulk samples. Bulk deposition thus includes wet deposition and a major proportion of dry deposition of trace elements.

Bulk deposition data were recorded following three criteria. First, the measurements were under good quality control (e.g., using standard samples, conducting recovery analysis). Second, bulk deposition was measured for more than a whole year during the period 2005–2021. Third, data were measured in the regions in the east of 'Hu Huanyong Line' where hold the majority of the national population and have experienced rapid urbanization (Chen et al., 2016). Data measured within mining areas and industrial estates were further excluded due to strong effects of local point source pollution. Bulk deposition of the eight trace elements was recorded using the same unit, i.e. mg $m^{-2}$ $yr^{-1}$, along with corresponding information on site locations (longitude and latitude), annual average temperature (°C), and annual precipitation (mm) for the sampling years from the literature. Data of multi-year measurements were averaged for the same site. Overall, our database included observed data from 108, 64, 132, 119, 111, 171, 124 and 150 sites for annual bulk deposition of Cu, Ni, Zn, As, Cr, Cd, Hg, and Pb, respectively (see Fig. S1 for locations of the sampling sties and Table S1 for raw data). Bulk deposition data were ln-transformed to fit a normal distribution and used for further statistical analysis.

#### 2.1.2. Distance and mediating factors

In China, the common pattern of urban development follows an expansion from the urban core to peri-urban areas, with higher population density and stronger human activities in more urbanized central areas (Li et al., 2021). We thus derived the distance (km) between the sampling site and the geometric center of the nearest large city (urban population $>$ 0.5 million) using Google Earth for Microsoft Windows (Version 7.1.5.1557, Google Inc. USA). The mining areal percentage (%) was calculated in an area with a radius of 100 km from the sampling site. Mining areal data were derived from the National Natural Resources and Geospatial basic information Database (http://sgic.net.cn/web/geo/zyzhjc/index.html.) to represent the anthropogenic emissions from mineral extraction. Industrial $SO_2$ emission ($10^4$ ton $yr^{-1}$) and industrial soot and dust emission ($10^4$ ton $yr^{-1}$) of the nearest large city for each sampling site were derived from China City Statistical Yearbook (2005–2021) (Department of Urban Surveys, National Bureau of Statistics of China, 2021) to indicate the anthropogenic emissions from coal combustion and related industrial activities.

#### 2.1.3. Urban soil concentrations of trace elements

We compiled a national database of urban topsoil concentrations (mg $kg^{-1}$) of eight trace elements (Cu, Ni, Zn, As, Cr, Cd, Hg, and Pb) using data both from literature and measured by our research group. The keywords 'trace element'/'heavy metal', 'urban soil' and China' were used to search target literature. To better match the database of bulk deposition, we recorded data on urban topsoil concentrations of trace elements following four criteria. First, the measurements were obtained by standard acid digestion preprocessing and assay methods (e.g., FAAS, ICP-AES, ICP-OES, ICP-MS). Second, good quality control was conducted using standard samples, duplicate samples and reagent blanks. Third, the literature was published during the period 2005–2021 and the data were measured from the regions in the east of 'Hu Huanyong Line'. Fourth, we only included topsoil (0–10 cm) data measured in green spaces or parks (mainly urban forests and lawns) from large cities with an urban population larger than 0.5 million. In addition, we collected surface soil samples from forest patches in 27 urban parks of nine large cities in eastern China (Guangzhou, Nanchang, Hefei, Zhengzhou, Shijiazhuang, Beijing, Shenyang, Changchun, and Harbin) during July–August 2019 (Wu et al., 2021). After standard preparation and acid digestion with a mixture of nitric, perchloric and hydrofluoric acids,

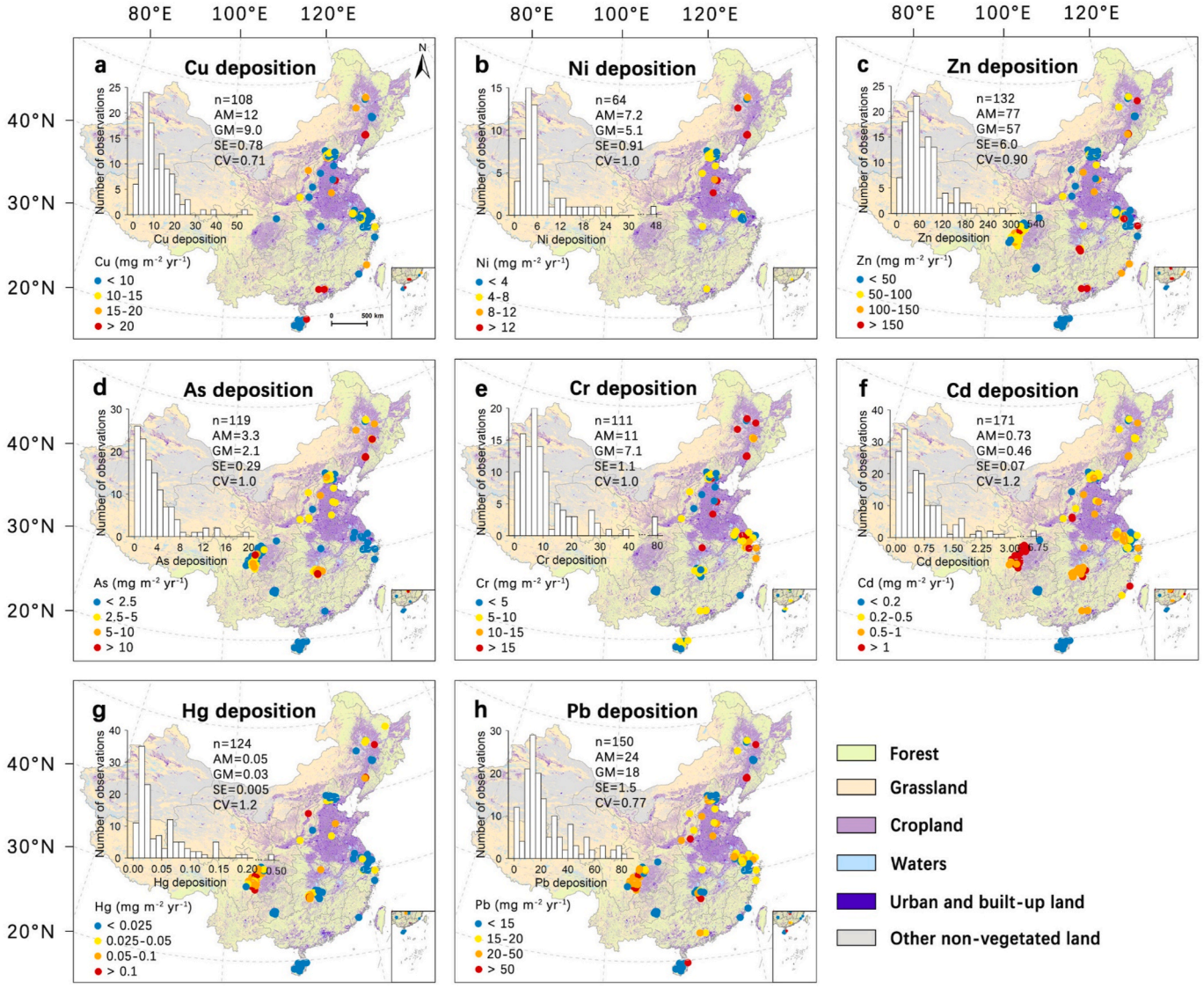


**Fig. 1.** Current bulk deposition (mg $m^{-2}$ $yr^{-1}$) of the eight trace elements in China. (a) Cu, (b) Ni, (c) Zn, (d) As, (e) Cr, (f) Cd, (g) Hg, and (h) Pb. Our analysis focuses on the regions in the east of Hu Huanyong Line where hold the majority of the national population and experiences rapid urbanization. AM and GM indicate arithmetic mean and geometric mean, respectively. Background shadows indicate the major land covers in China (Xu et al., 2018).

concentrations of Zn, Cr, As, Cu, Ni, Pb and Cd were measured using an inductively coupled plasma optical emission spectrometer (ICP-OES; Optima 5300 DV, PerkinElmer, the USA) and concentrations of Hg were measured using inductively coupled plasma source mass spectrometer (ICP MS, NexION 300X, PerkinElmer, the USA), respectively. Repeated measurements in the same city were averaged for further analysis. Overall, our database included observed data from 62, 44, 57, 47, 55, 55, 40 and 62 large cities for topsoil concentrations of Cu, Ni, Zn, As, Cr, Cd, Hg and Pb, respectively (Table S2).

### 2.2. Statistical and model analyses

#### 2.2.1. Testing urban hotspot hypothesis

To test the hypothesis of urban hotspot of bulk deposition, we first conducted a model selection analysis of five potential factors affecting bulk deposition (i.e., the distance between the sampling site and the geometric center of the nearest large city, annual precipitation, mining areal proportion, industrial $SO_2$ emission of the nearest large city, and industrial soot and dust emission of the nearest large city) based on Akaike's Information Criterion with a small sample correction (Calcagno and De Mazancourt, 2010). The importance of each predictor was estimated as the sum of the Akaike weights derived from model selection analysis and a cutoff value (0.8) was used to differentiate the important predictors (Calcagno and De Mazancourt, 2010). We further conducted conditional regression analysis of the relationship between ln-transformed bulk deposition of the eight trace elements with distance to the nearest large cities while holding all other important drivers constant. The multicollinearity of important drivers was assessed based on the variance inflation factor (VIF) where VIF $< 3$ indicates weak collinearity (Zuur et al., 2010). We estimated the variance explained by each important factor by averaging sequential sums of squares over all orderings of regressors (Groemping, 2006).

#### 2.2.2. Assessing current trace element pollution in urban soils

To evaluate the results of urban topsoil concentrations in terms of pollution levels, we collected data on national soil quality standards for domestic gardens, allotments, parks, playing fields, open space and recreational area and residential area of representative countries, including China, Canada, the USA, the Netherlands, and some other European countries (MEEC, 2018b; VROM, 2000; CCME, 1999; US EPA, 1994; ICRCL, 1987; See Table S3 for details). These soil quality standards are mainly defined based on potential risk to human health and

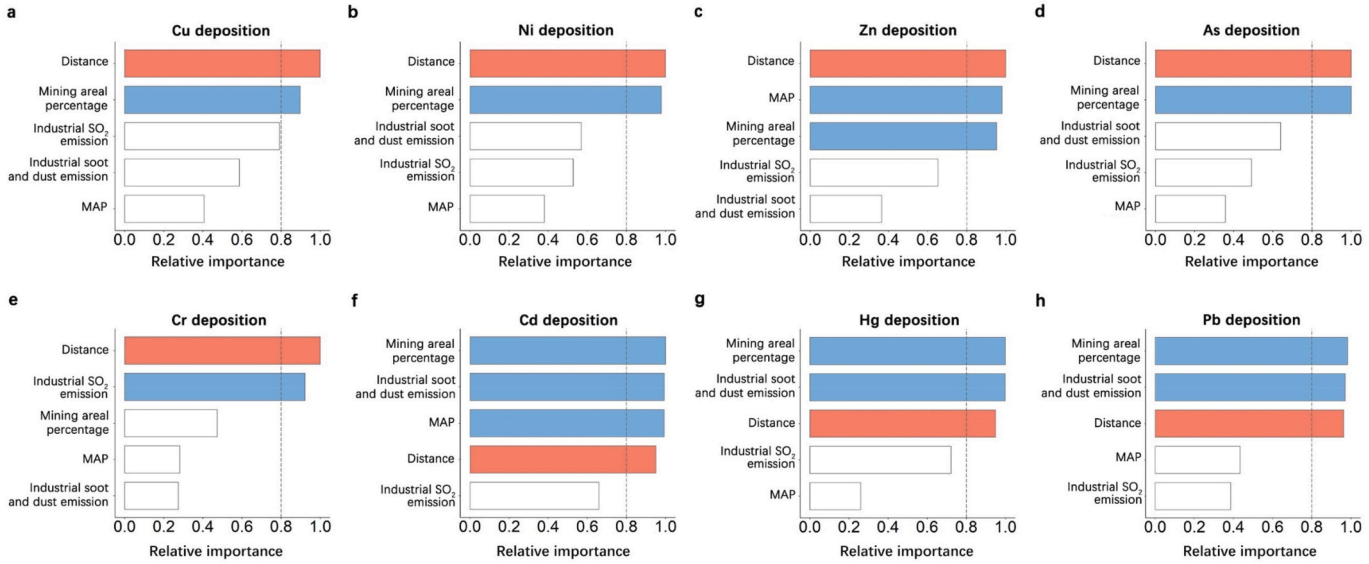


**Fig. 2.** Relative importance of the five potential predictors for the bulk deposition of the eight trace elements. (a) Cu, (b) Ni, (c) Zn, (d) As, (e) Cr, (f) Cd, (g) Hg, and (h) Pb. Distance to the urban core of the nearest large cities (in red) is among the most important predictors for all eight trace elements. Statistical analyses were based on ln-transformed values of bulk deposition. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

environmental sustainability, but the values of the standards vary in the various countries and consistent international standards are still lacking. We thus defined an urban soil quality standard as the mean of the 10% minimum values of all international standards of each element (i.e., 50 mg $kg^{-1}$ for Cu, 40 mg $kg^{-1}$ for Ni, 200 mg $kg^{-1}$ for Zn, 10 mg $kg^{-1}$ for As, 50 mg $kg^{-1}$ for Cr, 1.4 mg $kg^{-1}$ for Cd, 0.5 mg $kg^{-1}$ for Hg and 95 mg $kg^{-1}$ for Pb). We then calculated the proportion of the studied large cities with topsoil concentrations exceeding the urban soil quality standards.

### 2.2.3. *Assessing the contribution of bulk deposition to urban soil trace element pollution*

To further assess the contribution of bulk deposition to urban soil trace element pollution, we first tested the potential confounding impact of variations in background soil concentrations of the eight trace elements on the current values. We thus derived provincial soil background values of the eight trace elements (National Environmental Monitoring Centre of China, 1990) and tested their correlations with urban topsoil trace element concentrations.

After evaluating this potential confounding impact, a linear regression analysis was conducted to test the correlations between bulk deposition and urban topsoil trace element concentrations corrected by background values for natural soils (current values minus background values). To demonstrate the contribution of trace element deposition to soil pollution, we further conducted a scenario analysis of urban soil trace element concentrations, assuming that current levels of bulk

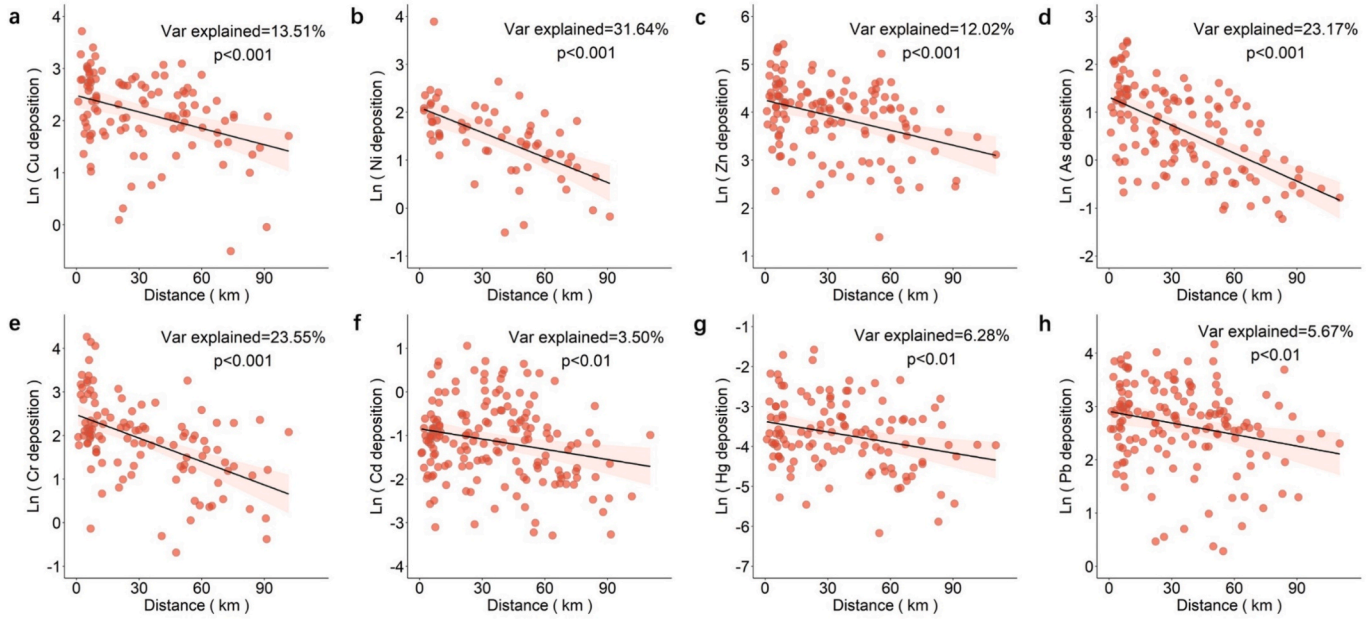


**Fig. 3.** Conditional regression plots showing significant increases in bulk deposition of the eight trace elements (mg $m^{-2}$ $yr^{-1}$) with closer distance to nearest large cities. (a) Cu, (b) Ni, (c) Zn, (d) As, (e) Cr, (f) Cd, (g) Hg, and (h) Pb. Statistical analyses were based on ln-transformed values of bulk deposition.

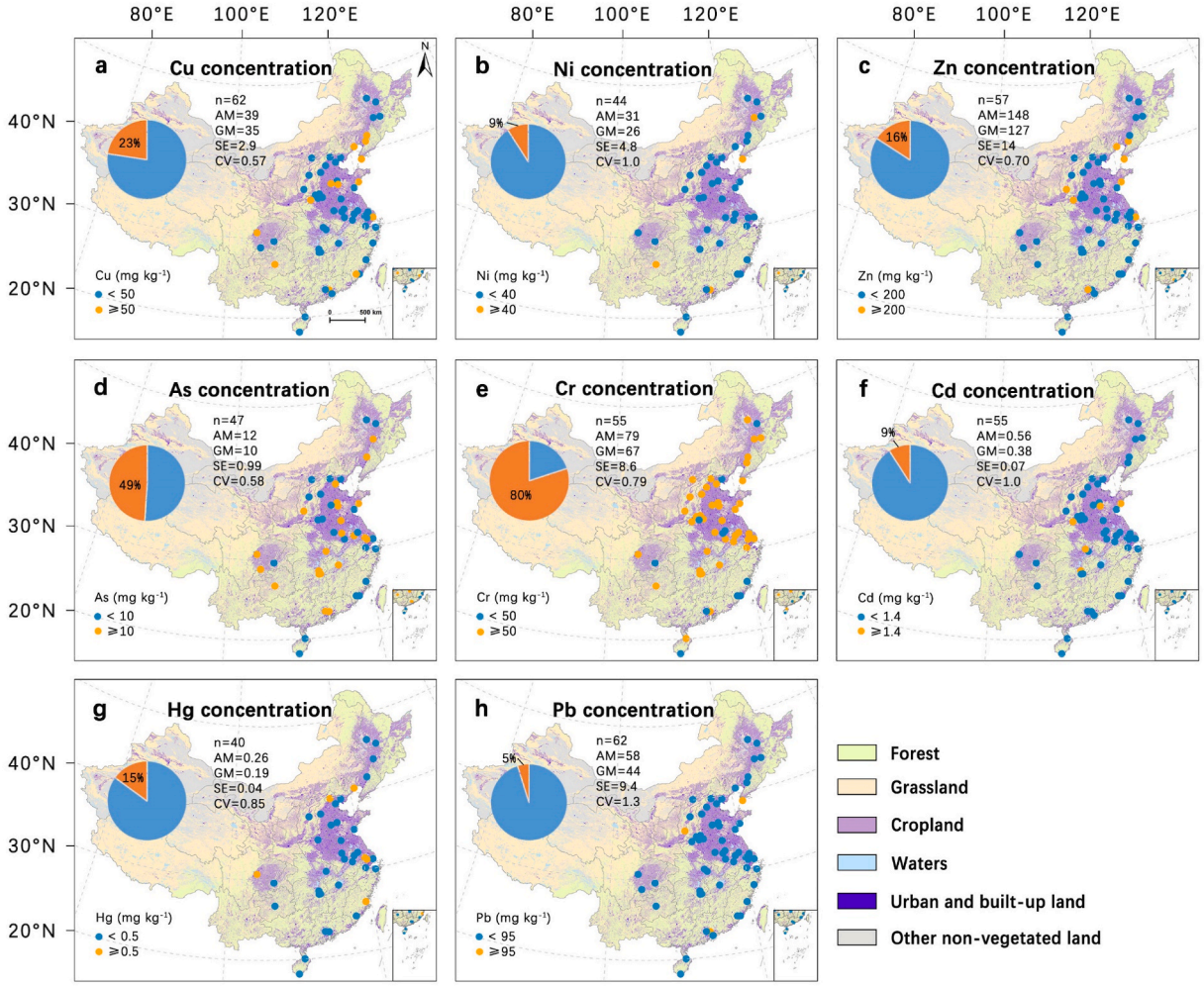


**Fig. 4.** Current urban topsoil concentrations of the eight trace elements (mg $kg^{-1}$) and their exceedances of urban soil quality standards. (a) Cu, (b) Ni, (c) Zn, (d) As, (e) Cr, (f) Cd, (g) Hg, and (h) Pb. The exceedance of the urban soil quality standards (See methods) for each trace element is indicated in orange in the inset pie plots, while values below the urban soil quality standards are indicated in blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

deposition will stay constant for twenty years. Assuming that the deposited trace elements accumulates completely in the topsoils (0–10 cm), we predicted topsoil concentrations of the eight trace elements twenty years later ($Con_1$, mg $kg^{-1}$) based on current topsoil trace element concentrations ($Con_0$, mg $kg^{-1}$), an average bulk density of urban surface soils (BD, 1.32 g $cm^{-3}$)(Table S4), and urban bulk deposition ($Dep_0$) (mg $m^{-2}$ $yr^{-1}$) according to Equation (1),

$$Con_1 = \frac{Con_0 \times BD \times 10 + Dep_0 \times 20}{BD \times 10} \quad (1)$$

Based on the predicted topsoil concentrations for each trace element, we further calculated the proportions of the studied large cities with topsoil concentrations exceeding the urban soil quality standards and compared them with current status. Spatial patterns of the bulk deposition and topsoil concentrations of the eight trace elements were illustrated using ArcGIS Desktop (version 9.3, ESRI, USA). All statistical analyses were performed using R software (version 3.6.3; R Development Core Team, 2020; http://www.r-project.org/).

## 3. Results

### 3.1. Spatial variations in bulk deposition of eight trace elements

Annual bulk deposition of the eight trace elements showed a decrease in the order Zn (geometric mean = 57 mg $m^{-2}$ $yr^{-1}$, ranging from 13 to 188 mg $m^{-2}$ $yr^{-1}$) > Pb (18, 2.7–66 mg $m^{-2}$ $yr^{-1}$) > Cu (9.0, 2.2–26 mg $m^{-2}$ $yr^{-1}$) > Cr (7.1, 0.9–30 mg $m^{-2}$ $yr^{-1}$) > Ni (5.1, 0.9–22 mg $m^{-2}$ $yr^{-1}$) > As (2.1, 0.5–11 mg $m^{-2}$ $yr^{-1}$) > Cd (0.46, 0.09–2.2 mg $m^{-2}$ $yr^{-1}$) > Hg (0.03, 0.01–0.15 mg $m^{-2}$ $yr^{-1}$) (Fig. 1; Table S1). Bulk deposition of As, Cr, and Ni increased significantly towards the east ($p < 0.01$), while bulk deposition of Cd showed an opposite trend ($p < 0.001$) (Fig. S2). Additionally, there was a significant increase in bulk deposition of As and Cr towards the north ($p < 0.001$), while bulk deposition of Cd showed an increase towards lower latitudes ($p < 0.001$) (Fig. S3).

Model selection analyses showed that the distance to the nearest large cities was among the most important predictors for the spatial variation in bulk deposition of all the eight trace elements (Fig. 2). In line with our urban hotspot hypothesis, conditional regression analyses demonstrated that the ln-transformed bulk deposition of the eight trace elements all showed a significant increase with closer distance to the nearest large cities (variance explained 14% for Cu, 32% for Ni, 12% for Zn, 33% for As, 24% for Cr, 3.5% for Cd, 6.3% for Hg and 5.7% for Pb, respectively; Fig. 3; Tables S5–S12). This implies an exponential increase in the bulk deposition of the eight trace elements towards the large cities.

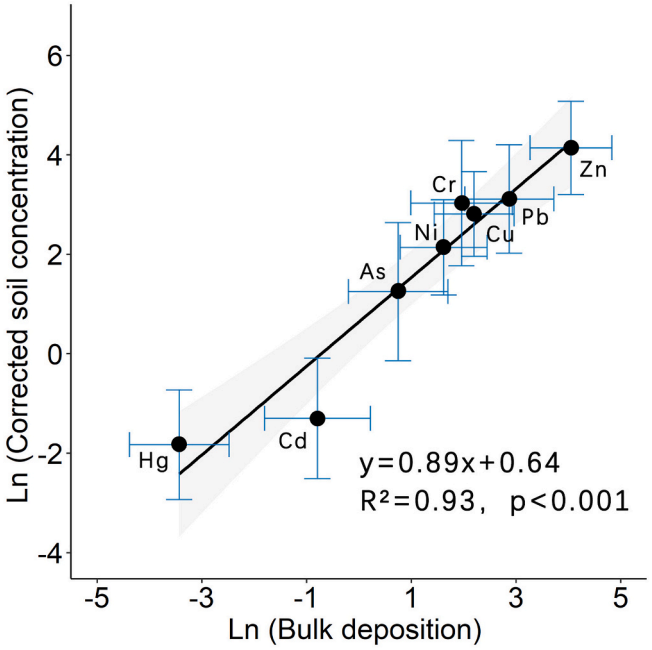


**Fig. 5.** Relationship between urban topsoil concentrations of trace elements (mg $kg^{-1}$), corrected by background values for natural soils, and urban bulk deposition (mg $m^{-2}$ $yr^{-1}$). Statistical analyses were based on ln-transformed values. Error bars indicate standard errors of ln-transformed values.

### 3.2. Factors mediating urban hotspots of trace element deposition

Urban hotspots of bulk deposition of the eight trace elements were found to be mediated by both anthropogenic variables and climatic variables (Fig. 2). Specifically, bulk deposition of Cu, Ni, Zn, As, Cd, Hg and Pb increased significantly with higher mining areal percentage (variance explained 3.7% for Cu, 14% for Ni, 4.8% for Zn, 9.7% for As, 12% for Cd, 7.2% for Hg and 6.6% for Pb, respectively; Tables S5–S8, Tables S10–S12). Bulk deposition of Cd, Hg and Pb increased significantly with higher industrial soot and dust emission (variance explained 3.5% for Cd, 9.8% for Hg and 7.5% for Pb, respectively; Tables S10–S12). Bulk deposition of Cr increased significantly with higher industrial $SO_2$ emissions, a rough surrogate of coal combustion (variance explained 4.9%; Table S9). Additionally, bulk deposition of Zn and Cd increased significantly at higher annual precipitation (variance explained 4.3% for Zn and 5.9% for Cd, respectively; Tables S7 and S10).

### 3.3. Potential effects of bulk deposition on topsoil trace element pollution

Urban topsoil concentrations of the eight trace elements followed the order Zn (geometric mean = 127 mg $kg^{-1}$, ranging 54–270 mg $kg^{-1}$) > Cr (67, 31–162 mg $kg^{-1}$) > Pb (44, 20–95 mg $kg^{-1}$) > Cu (35, 18–81 mg $kg^{-1}$) > Ni (26 mg $kg^{-1}$, 12–42 mg $kg^{-1}$)> As (10, 5.2–24 mg $kg^{-1}$) > Cd (0.38, 0.10–1.8 mg $kg^{-1}$) > Hg (0.19, 0.06–0.68 mg $kg^{-1}$) (Fig. S4; Table S2). Currently, urban topsoil concentrations exceeded the urban soil quality standards in 80% of the studied cities for Cr, 49% for As, and below 25% for other trace elements, respectively (Fig. 4). Current bulk deposition of trace elements within 20 km to the central urban area was in the order Zn (geometric mean = 76 mg $m^{-2}$ $yr^{-1}$, ranging 25–238 mg $m^{-2}$ $yr^{-1}$) > Pb (23, 8.5–75 mg $m^{-2}$ $yr^{-1}$) > Cu (12, 5.0–33 mg $m^{-2}$ $yr^{-1}$) > Cr (11, 3.3–46 mg $m^{-2}$ $yr^{-1}$) > Ni (9.7, 4.5–25 mg $m^{-2}$ $yr^{-1}$) > As (3.7, 0.7–14 mg $m^{-2}$ $yr^{-1}$) > Cd (0.5, 0.1–2.5 mg $m^{-2}$ $yr^{-1}$) > Hg (0.04, 0.01–0.15 mg $m^{-2}$ $yr^{-1}$). Urban soil concentrations of most trace elements (except As and Hg) showed no significant correlations with background values for natural soils (Fig. S5). After a correction by the background values for natural soils, urban topsoil concentrations of trace elements showed a significant increase with corresponding bulk deposition to urban areas ($R^2 = 0.93$, $p < 0.001$; Fig. 5). Assuming that the urban bulk deposition of trace elements stay constant in next twenty years, we predicted that urban surface soil concentrations would exceed the urban soil quality standards in 100% of the studied cities for As, 96% for Cr, 95% for Zn, 73% for Ni, 69% for Cd, 69% for Cu, 31% for Pb, and 18% for Hg, respectively (Fig. 6).

## 4. Discussion

### 4.1. Urban hotspots of atmospheric trace element deposition

In line with the urban hotspot hypothesis, bulk deposition of the eight trace elements showed a significant increase with proximity to the nearest large cities. Distance to the urban core is a surrogate of multiple anthropogenic drivers that may increase the atmospheric deposition of trace elements. First, trace elements can be emitted from construction activities, urban building weathering, and hard road wearing and consequently deposited in the urban environments (Zhao et al., 2011). Second, the use of gasoline type fuels, tire and brake pad wear, oil and lubricant can contribute significantly to emissions and deposition of Cu, Zn, Cd and Pb (Harrison et al., 2012; Huber et al., 2015), while Cr and Ni from the wear of metallic parts and chrome accessories may lead to higher deposition in urban areas (Yan et al., 2022). Third, fossil fuels are widely used in urban heating, cooking and industry processes, emitting large amounts of As, Hg and other trace elements that further enhance regional deposition of these elements (Zhao et al., 2011; Li et al., 2015). Moreover, burning treatment of urban waste is also an important source that increases atmospheric deposition of trace elements (e.g., Cu, Zn, As, Cr and Pb) in and around large cities (Cheng et al., 2020).

Anthropogenic point source emissions can significantly mediate the urban hotspots of trace elements. First, we found that mining activities significantly increased the bulk deposition of Cu, Ni, Zn, As, Cd, Hg and Pb deposition. China has abundant mineral resources and the mining activities can cause dust emissions of trace elements (Li et al., 2014; Schlesinger et al., 2022), thereby enhancing atmospheric deposition to surrounding areas. Many trace elements often co-occur in mineral resources due to their geochemical nature (Adrianto et al., 2022) and this explains the significant increase in bulk deposition of various trace elements with the distance to mining areas (Tables S5–S8, Tables S10–S12). Second, our results show that bulk deposition of Cr increased significantly with industrial $SO_2$ emission which is a surrogate of the combustion of sulfur-containing fuels (e.g., coal) that are important sources of Cr emissions (Yan et al., 2001; Stam et al., 2011). Third, bulk deposition of Cd, Hg and Pb were found to increase with industrial soot and dust that comes from mechanical crushing and grinding of solid materials, the mixing, sieving, packaging and transport of powdery materials, as well as the soot generated by the combustion (Wang et al., 2013; Duan and Tan, 2013). Additionally, our results indicate a significant increase in bulk deposition of some trace elements (e.g., Zn and Cd) with higher annual precipitation, implying that the spatial variation in precipitation may also modify the urban hotspots of atmospheric deposition.

### 4.2. Potential urban soil pollution risks from trace element deposition

Our results show that concentrations of trace elements in urban topsoils are on average 1.1 to 5 times of the average background values for natural soils in China, being estimated as the national geometric mean values of more than 4000 background sites unaffected by pollution, mining activity, and traffic sources (Chen et al., 1991). Specifically, urban topsoil concentrations exceeded the urban soil quality standards in 80% of the studied cities for Cr, 49% for As, and less than 25% for other trace elements. We further found a significant increase in corrected urban topsoil concentrations of trace elements with corresponding bulk deposition to urban areas, suggesting that high-level atmospheric inputs can potentially contribute to the accumulation of

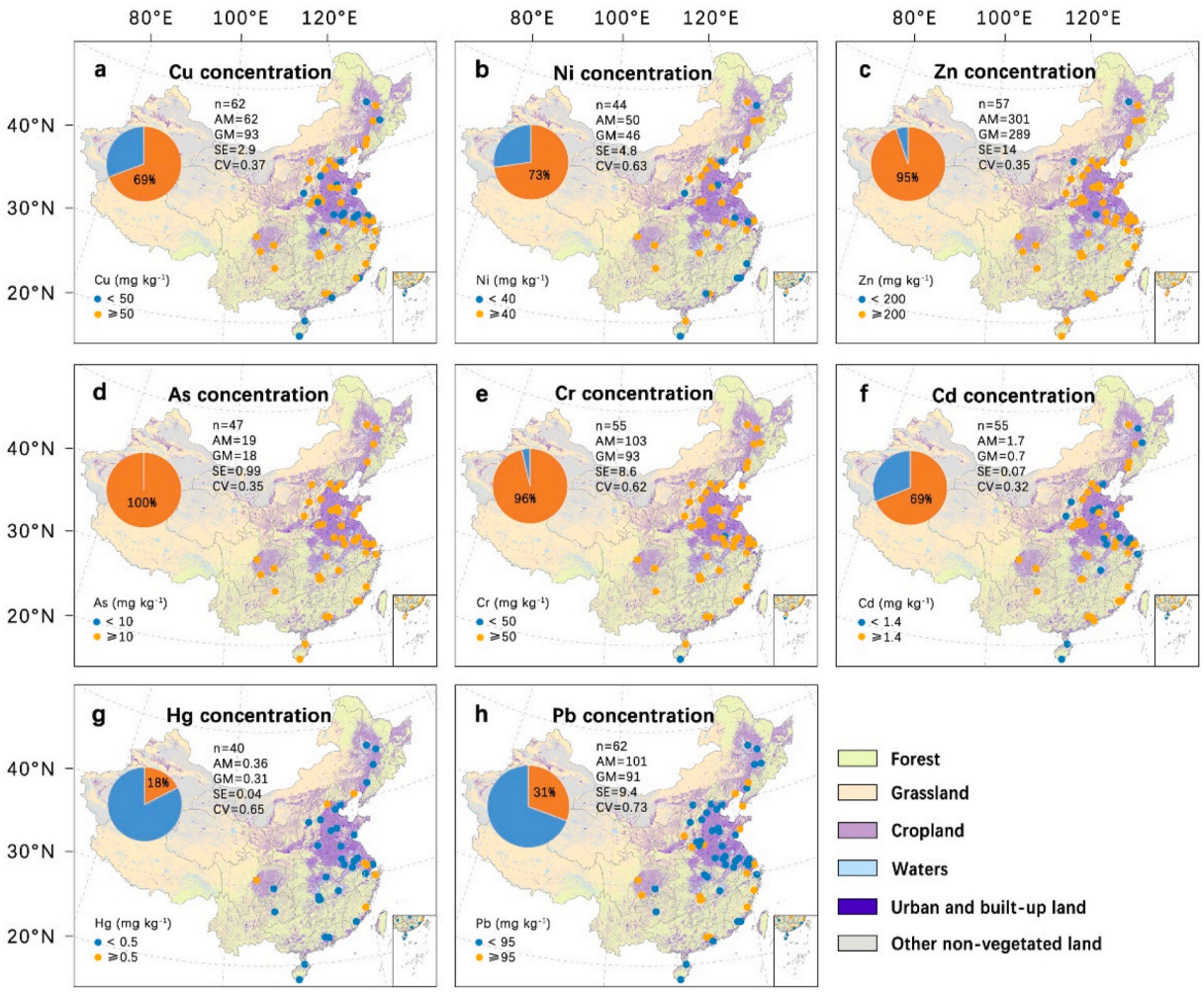


**Fig. 6.** Predicted urban topsoil concentrations of the eight trace elements (mg $kg^{-1}$) and their exceedances of urban soil quality standards assuming that bulk deposition of trace elements stay stable in twenty years. (a) Cu, (b) Ni, (c) Zn, (d) As, (e) Cr, (f) Cd, (g) Hg, and (h) Pb. The exceedance of the urban soil quality standards (See methods) for each trace element is indicated in orange in the inset pie plots, while values below the urban soil quality standards are indicated in blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

trace elements in urban surface soils. Assuming constant levels of bulk deposition in twenty years, a scenario analysis demonstrated that the proportion of the studied cities with exceedance of the urban soil quality standards would increase dramatically (i.e., > 95% of the studied cities for As, Cr, and Zn; > 65% for Ni, Cd, and Cu), thereby confirming the significant contribution of atmospheric deposition to urban soil trace element pollution.

Our findings highlight a threat of high-level urban trace element deposition to the soil environment against the goals of sustainable development of cities. Urgent measures are thus needed to cut atmospheric deposition of harmful trace elements in China's urban areas with a specific focus on Cr and As. Currently, emission control policies have been gradually implemented in Chinese cities, including energy transition, closing small and medium-sized coal-fired boilers, phasing out excess production capacity such as steel and coking, promoting the use of new energy vehicles, and strengthening dust control (Bo et al., 2021; Zheng et al., 2016). These measures promise a future reduction of atmospheric trace element deposition to some extent but stricter policies for trace element pollution control still need to be implemented.

### 4.3. Uncertainties and limitations

Several uncertainties remain in our analyses. First, bulk deposition is likely an underestimate of the total deposition for urban forests and parks. Total deposition of heavy metals to forest can be substantially higher than the bulk deposition to open land because of the higher surface roughness. For instance, the ratios of total deposition at forested sites to bulk deposition at nearby non-forested sites in the Netherlands were estimated to be 2.6 for Cu, 2.4 for Zn, 2.0 for Ni, 2.9 for Cd, 1.2 for Cr, and 2.3 for Pb, respectively (De Vries and Bakker, 1998). Although the ratios may differ for China, the total deposition of trace elements was likely underestimated in our analysis. Second, the parental material is an additional factor affecting the concentration of trace elements in soils. However, we found no significant correlations between urban soil concentrations of most trace elements (except As and Hg) with background values for natural soils, suggesting the importance of other external inputs instead of pedogenic sources for these trace elements in urban soils. Our correlation analysis with bulk deposition was reasonable as we used values of urban topsoil concentrations of trace elements corrected by background values for natural soils. Third, we evaluated the urban hotspot hypothesis using the distance between the sampling site and the geometric center of the nearest large city (Du et al., 2015, 2018) in view of a common pattern of urban expansion from the urban core to peri-urban areas in China (Li et al., 2021). In some cases, the geometric center of a city is not necessarily the most populated area due to the spatial pattern of topography and water systems. This may potentially cause uncertainties in our analysis of the urban hotspot hypothesis.

Limitations of available datasets can also lead to uncertainty in our

study. The time of measured data spanned more than ten years for both bulk deposition and urban topsoil concentrations of trace elements. The implementation of air pollution control policies (e.g., Air Pollution Prevention and Control Action Plan in 2013) may lead to a change of atmospheric deposition over time, but the temporal variations of bulk deposition and soil concentrations of trace elements were not considered in our statistical analysis. Urban soils may also receive external inputs of trace elements from other sources, including fertilization and irrigation. However, these potential sources are not included in our analysis due to a lack of data.

Our scenario analysis neglected on one hand the input of trace metals by dry deposition and on the other hand the plant uptake of micronutrients and leaching of trace metals to deeper soils. First, bulk deposition is likely an underestimate of the total deposition for urban forests and parks (De Vries and Bakker, 1998). Inversely, we do not account for trace metal uptake and leaching while either one or both terms can counteract the neglected dry deposition of trace metals. For example, some of the trace elements are also micronutrients (e.g., Cu, Ni and Zn) that can be utilized by plants (Clarkson et al., 1980). Leaching to deeper soils is very limited for Pb, Cu and Hg which form strong complexes with organic matter (Bergkvist et al., 1989), but it can be substantial for Cd and Zn (Bergkvist et al., 1989; Pedroli et al., 1990; Wilkens, 1995). Overall, the impact of neglecting dry deposition is likely larger for most trace elements than neglecting uptake and leaching, with the possible exception of Cu and Zn, implying that the calculated risks are indicative for the deposition impacts on urban soil quality. Future research efforts are needed to address the abovementioned uncertainties for better understanding of urban topsoil trace elements.

## 5. Conclusions

Our results confirm the occurrence of urban hotspots of atmospheric trace element deposition in China. The bulk deposition of trace elements increased significantly with closer distance to the large cities, implying that urban areas are concentrators of trace elements. We further highlight the high pollution risks of Cr and As in urban surface soils. High-level atmospheric deposition has a potential to cause a substantial increase in the exceedance of urban soil quality standards for the trace elements. Moreover, it should be noticed that trace elements are not only enriched in the soils, but also may enter urban groundwater directly through atmospheric deposition (Mcmahon et al., 2019). The urban hotspots of atmospheric trace element deposition can also result in high-level inputs of trace elements to the farmland belts surrounding large cities (Meharg and Meharg, 2021). Overall, timely efforts are needed to reduce urban emissions of trace elements and alleviate negative effects on regional soil environment and human health for a sustainable future of cities in China.

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## CRediT authorship contribution statement

**Yuying Guo:** Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **Enzai Du:** Conceptualization, Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization, Supervision. **Binghe Li:** Investigation, Data curation, Writing – original draft. **Nan Xia:** Investigation, Data curation, Writing – original draft. **Xinhui Wu:** Investigation, Data curation, Writing – original draft. **Wim de Vries:** Formal analysis, 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

Data will be made available on request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.137872.

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