Pol. J. Environ. Stud. Vol. 32, No. 5 (2023), 4113-4128
DOI: *10.15244/pjoes/166355* ONLINE PUBLICATION DATE: 2023-08-09

Original Research

Trace Metal Content, Ecological Risk Assessment in Forest Soils and Correlation with Accumulated Gross Domestic Industrial Product in the City of a Highly Industrialized Area, Dongguan, China

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Received: 23 March 2023
Accepted: 20 May 2023

Abstract

To evaluate the pollution and ecological risk of trace metals in the forest parks of highly industrialized manufacturing Cities, soil from three different depths and plant root samples were collected in December 2020 from six major forests of Dongguan, China. The level of pollution in seven metals is decreasing in the following order: Zn>Cr >As>Pb>Cu>Ni>Cd. Elemental spatial and vertical distribution show that the degree of urbanization influences the extent of pollution. Moreover, the trace metals were intercepted at the soil's surface or sediment layer, becoming absorbed by roots of pines, trees, and ferns. The potential ecological risk of the metals was moderate-to-high ecological risk rank and followed the order: Cd>As>Cu>Ni>Pb>Cr>Zn. Source apportionment indicated that the Cd, As, Cu, Pb, and Zn might have originated from industrial activities, while Ni and Cr from natural sources. The amount of total metals in soils from forest were significantly associated with the accumulated twenty years' gross industrial product (GDP) for secondary industries values and also the history of rapid industrialization. This paper reported for the first time the correlation between heavy metal pollution in forest soils and gross industrial product.

Keywords: urban forest, soil pollution, trace metals, ecological risk, bio-enrichment

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Introduction

Urban forests, often regarded as "the lungs of the city", are essential natural filters for air pollution, thereby refreshing the air [1]. It can also reduce noise pollution, alleviate urban thermal pollution, and improve the microclimate [2]. Therefore, as part of green infrastructure in the city, urban forests are significant for sustainable urban environments. However, due to their location in the human-gathering area, urban forest ecosystems are more vulnerable than natural forest systems [3]. Recently, soils in urban forests are deteriorating due to the increasing assimilation of potentially harmful substances from urban development [4]. The rising urban population also burdens the urban forests with potentially toxic elements. Among different pollutants, trace (toxic) metals are more harmful to human health, either directly or indirectly [5], because of their high toxicity, non-biodegradability, persistence, and bioaccumulation in the food chain [6]. Therefore, the environmental risk evaluation of trace metal-contaminated soils is of substantial public concern in recent decades. However, most previous studies focused on large industrial areas, rural cultivated land areas, and urban soils [7-8], while limited information is available on trace metal pollution of soils in urban forests.

Trace metals in forests originate from both natural and anthropogenic sources. The major natural source is rock weathering. Others include active volcanoes, earthquakes, forest fires, and other processes that suspend the metals in the air and later enter the soils by wet or dry precipitation. However, trace metals consistently enter the environment through anthropogenic activities, such as agricultural and industrial processes, electronic wastes, wastewater irrigation, and indiscriminate waste disposal, leading to the deterioration of soil quality [9-10]. For example, electroplating wastewater usually contains various metals, including Cr, Ni, Cd, Cu, and Zn [7]. Besides, boiler combustion can emit Pb, Cr, and Cu attached to particles [11]. Elsewhere, mining is a primary anthropogenic source for metal pollution of soils [12]. Cd and Zn concentrations near mining sites are much higher than those in the control area [13].

Trace metal pollution of forest soil can pose severe threats to cultivated soils, plants, vegetables, and food chains, causing human health-related hazards [14]. Trace metals in soil can bioaccumulate in plants and different organs of the human body through ingestion, causing acute or chronic poisoning [5, 15]. Long-term exposure to Cd can cause health issues, such as lung cancer, hypertension, kidney dysfunction, and bone fractures [16]. For instance, Cr with different valency is highly toxic in the human body [17]. Moreover, trace metals alter the circulation of nutrients and change the soil ecosystem by potentially changing the metabolic activity of soil organisms [18], thereby reducing their diversity. Several trace metals negatively impact the genome of organisms, causing chromosome aberration and DNA strand breaks. Excessive Cu in soils retards plant growth and causes leaf chlorosis [19]. Moreover, different forms of trace metals have different migration rates and bioavailability in soil, which determines their complex behaviors and ecological effects in the environment [20]. Therefore, it is of great significance to understand the spatial distribution pattern of trace metals in soil and carry out ecological risk assessment of trace metals in soil and living organisms in forest areas for the control and treatment of metal pollution.

Dongguan, known as "the world's factory", is one of the most crucial industrial cities in the Guangdong-Hong Kong-Marco Greater Bay Area. In recent three decades, manufacturing industries, including clothing, electronic, electrical machinery, furniture, toys, hardware, and other industries, have been prosperously developed. These industries have been integrated into the international market [21]. However, the exponential industrialization and urbanization of Dongguan have resulted in widespread soil contamination by trace metals from different anthropogenic sources [22]. For instance, As and Cr have been found in their topsoils and have exhibited potential health risks [22]. With the continuous emission of trace metals from these factories, the soils are susceptible to higher ecological risk [22]. Dongguan has 15 forest parks, and the environmental problems caused by trace metal pollution require diligent investigation. Therefore, the present study aims to (1) determine the content, distribution, and bioaccumulation of trace metals in Dongguan urban forest parks, (2) assess the pollution status and ecological risk of forest soils using various pollution indexes, (3) carry out source apportionment for the metals in the soil using multivariate statistical techniques, (4) metal content with the degree of industrialization.

Materials and Methods

Study Area and Sample Collection

The Dongguan City (22°39'-23°09'N, 113°31'-114°15'E), located in the center of Guangdong-Hong Kong-Macao Greater Bay Area, is an internationally renowned manufacturing base. Dongguan has a subtropical monsoon climate with average annual temperature of 23.3ºC and annual rainfall of 1820 mm. Dongguan has experienced rapid economic and social development in these decades. However, the exponential industrialization and urbanization of Dongguan have resulted in widespread soil contamination by trace metals from different anthropogenic sources, and the soils are vulnerable to higher ecological risk. Dongguan has 15 forest parks, and the environmental problems caused by trace metal pollution require diligent investigation.

The soil and plant (pines, arbors, ferns) root samples were collected in the urban forest park of Dongguan City. The sampling was designed to compare the distribution of trace metals in the forest soils with distinct urbanization and industrialization gradients. Based on the population density and the division of active areas, the forests were divided into three sampling areas: the urban central area (Huangqishan City Park (HQS) and Tongsha Ecological Park (TS)), urban peripheries (Shuilianshan Forest Park (SLS) and Dalingshan Forest Park (DLS)), and suburban area (Dapingzhang Forest Park (DPZ) and YinpingshanForest Park (YPS)). Furthermore, the number of sampling points was determined by the sampling area size (Fig. 1). Two, three, and four sampling points were selected for small-size (HQS City Park, TS Ecological Park, SLS Forest Park, and DPZ Forest Park), medium-size (DLS Forest Park), and large-size (YPS Forest Park) sampling areas, respectively.

The surface soil samples were collected using a hand-driven stainless-steel soil auger. Five soil samples from each sampling site were homogenized into a composite (100 g). In addition, stratified sampling on the vertical section of each plane was done. Stratified soil samples at 0-20, 20-40, 40-60 cm depths were collected with a spatula (Fig. S1). Composite samples from the same layer were also homogenized (100 g). Meanwhile, plant roots (50 g) of tall arbors (above 2 m), pines, and ferns were also collected at each sampling site [23].

Sample Pretreatment and Trace Metal Analysis

The collected roots were copiously rinsed with deionized water before air-dried under drafty conditions. Meanwhile, stones and organism debris were removed from the soil samples and turned at regular intervals. After drying, the soil samples were filtered through 0.15 mm mesh, whereas the roots were cut into ≈10 mm by clean ceramic scissors.

Respectively, 0.10 and 0.20 g of the soil and root samples were digested by 8 mL aqua regia (HNO_3/HCl, 1:3, v/v). Afterward, the mixture was placed in a Teflon-sealed tank, which enables microwave digestion. After cooling, the digested mixture was diluted with deionized water to 50 mL. A sample blank (control) was also prepared.

Furthermore, the concentrations of Cu, Zn, Ni, Cr, Cd, Pd, and As in soil were determined by inductively-coupled plasma mass spectrometer (ICP-MS, Agilent 7500cs, USA). We used the soil environment quality risk control standard for soil contamination of agricultural land (GB15618-2018) to characterize the metals. The background value was that of Guangdong Province [24].

Pollution and Risk Assessment

To comprehensively evaluate the trace metal contamination of the soils, we applied pollution indices (i.e., geo-accumulation index (Igeo), enrichment factor (Ef), Nemerow's pollution index (PI), biological enrichment factor (BAF), biogeochemical Index (BGI), potential ecological risk index (RI), and Modified pollution index (MPI)) for environmental and ecological risk assessment [25]. Moreover, we related the Igeo

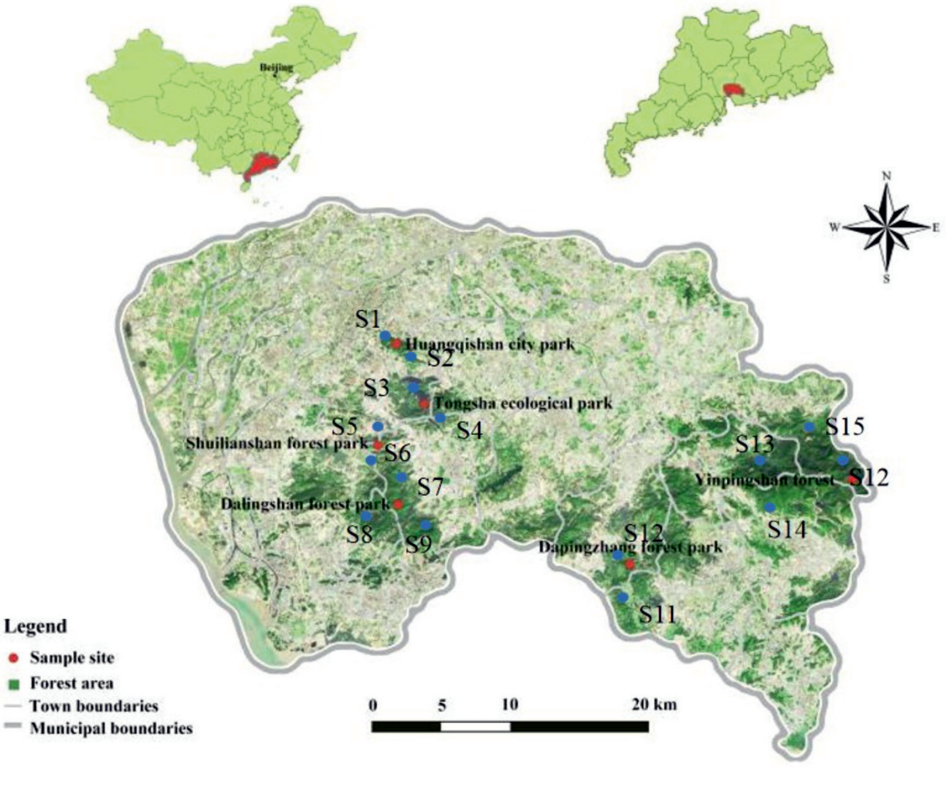

Fig. 1. Map of the study area showing sampling locations.

and Ef of the metals and their soil concentrations to the PI. The MPI reflects the soil pollution condition by incorporating metals' bio-availability, while BAF indicates the metal concentration in the soil organisms' body. The RI evaluates the potential ecological risk. Detailed definition and expression of these pollution indices are provided in Table S1.

Statistical Analysis

Further, we used SPSS 20 to analyze the differences and correlations between the trace metals and the indicators of co-existing trace metals being of the same source. Pearson's correlation coefficient investigated the relationship between the two variables; the larger the coefficient, the stronger the correlation. Additionally, principal component analysis (PCA) linearly converted multiple variables into fewer essential ones (i.e., principal components). These new variables could replace and reflect the original information through dimension reduction. Moreover, linear regression analysis were also adopted to test the correlation between metal content in soil and the gross domestic product for secondary industries of regional area.

Results and Discussion

Soil Characteristics and Statistical Analysis of Trace Metal Distribution

The physico-chemical characteristics of the soils were summarized in Table 1. The pH of the soils ranged from 5.66 to 6.10, with a slight pH rise with increasing soil depth. The moisture content of the soil ranged from 23.53 to 28.19. The moisture content also raised slightly with increasing soil depth. In addition, the content of organic carbon, total nitrogen and total phosphorus were in the range of 13.60-17.16, 0.64-1.14 and 1.16-1.19, respectively.

The coefficient of variation (CV) reflects the average degrees of variation between samples. Generally, CV≤0.1, 0.1<CV<1, and CV≥1 indicate weak, moderate, and strong variabilities, respectively. A large CV indicates external interference [5]. For the soil samples, the CV range for Cr, As, Ni, and Zn in each soil layer 0.19-0.68, inferring the influence of geological factors (Fig. 2). However, with respective CVs of 0.63-1.03, Cd and Pb were influenced by both geological and anthropogenic factors, whereas Cu with the CV of 0.81-1.5 indicates anthropogenic-driven pollution.

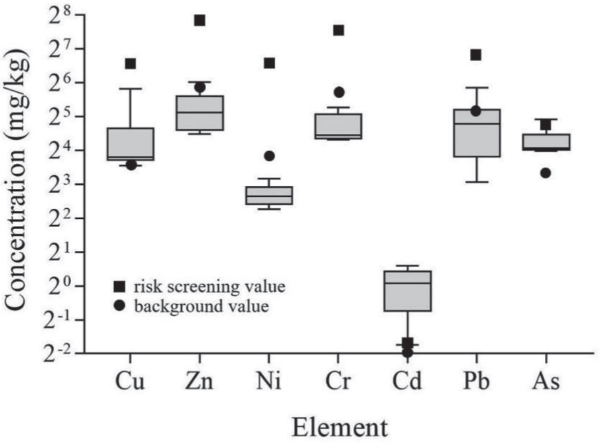

Fig. 2. Trace metal concentrations in the surface layer of the forest soil samples.

The CV values of trace metals in the root samples were >0.1, suggesting moderate-to-strong degrees of variation. Apart from the strong variability of Cd and Pb, others in the fern roots were moderate. Cu, Zn, and Cd demonstrated strong variation regarding the pine roots, while Ni, Cr, Pb, and As were moderately varied. Except for Pb's strong variability (CV = 1.12) in the arbors, other metals evinced CV of 0.70-0.96 (Table S2), indicating moderate variations. Generally, the concentration and distribution of the trace metals in the plant roots suggest that human activities predominantly caused pollution.

Distribution of the Trace Metals in the Soils

The median concentration ranges of the trace metals in soil (mg/kg) follow the order: 0.25-3.64 Cd<2.67-11.6 Ni<11.2-140 Cu<7.68-107 Pb<11.2-66.3 As<14.0-49.0 Cr<19.0-110 Zn (Fig. 2). Compared with the soil standard (GB15618-2018), the Cd content exceeded the risk screening value, indicating that the soil ecological environment might be in danger. In addition, the Cd content of the soils was 5.88 times than that of New Zealand's forests, which is considered "green" [26]. The influence of nearby industrial activities was suspected for the high Cd concentrations.

The median concentrations of Cu, Zn, Ni, Cr, Cd, Pb, and As in the study area's surface soil were 13.4,

Table 1. Physico-chemical characteristics of forest soil samples.

Depth (cm)	pH	Moisture content (%)	Organic carbon (g/kg)	Total nitrogen (g/kg)	Total phosphorus (g/kg)
0-20	5.73±0.07	25.04±1.51	17.16±2.13	0.91±0.08	1.19±0.17
20-40	5.86±0.06	25.85±1.70	14.52±1.59	0.64±0.03	1.16±0.09
40-60	6.02±0.08	26.44±1.75	13.60±1.51	1.14±0.09	1.74±0.11

25.5, 6.02, 21.3, 0.75, 13.6, and 15.7 mg/kg, respectively. Therefore, Cu, Cd, and As were 1.85, 15.16, and 1.89 folds of the Guangdong Province's background values [27]. Additionally, S4 had the highest content of Zn and Pb, attributable to the numerous tourists often encountered at the Tongsha Ecological Park and the automobile exhaust in the vicinity. Nevertheless, the highest contents of Cu, Cd, and As were observed at S11 of the Dapingzhang Forest Park, likely originated from the excessive use of chemical fertilizers and pesticides containing heavy metals, such as fosfomycin, manganese zinc, zinc difenacoum, etc. Furthermore, Cu (CV = 1.46) and Pb (CV = 1.07) both have strong variability, indicating the influence of the external environment.

To further analyze the degrees of pollution, we compared our current results with those of previous studies. The higher median levels of Cu, Zn, Ni, Cr, Pb, and As in the forest around Dexing copper mine (in Jiangxi province) were higher than those in the present study area [28], likely due to the mining activities. Also, Zn, Ni, and Cr concentrations in our study were lower than those in Shanghai, while Cu was mildly higher [29]. Compared with soil samples in an industrial region of Auckland, New Zealand, the levels of Zn, Ni, and Pb in our soil samples were lower, whereas those of Cr and Cd were higher [26]. Moreover, aside from Cd and As, the concentrations of other trace metals in the urban topsoil of Dongguan were excessively higher than those in the study area [30]. Generally, we found Cu and Cd at moderate-to-severe pollution levels.

Usually, trace metals adsorbed onto the surface layer of soils. Their vertical distribution in the soil is driven primarily by the soil's physicochemical properties and human activities [31]. As expected, the concentrations of Cd, Pb, and As decrease with depth, indicating that the metals, with low percolation power, concentrate on the topsoil. However, Cu, Zn, and Cr were most concentrated in the sediment layer (depth of 20-40 cm) (Fig. S1). Moreover, the total trace metal concentration was highest in the surface layer, attributable to human activities and natural sources. By comparison, the total concentrations of Zn, Ni, Cr, and Pb in the surface soil samples were lower than the background values [32]. Therefore, the vertical distribution of these metals might be driven by the soil's physicochemical properties or the elements' migrational ability. Contrarily, the contents of Cu, Cd, and As in the surface layer were higher than the background value, probably due to human activities including industrial metal processing, machinery manufacturing, and use of metal-containing pesticides and fertilizers.

Metal Concentration in the Root Samples

The concentration ranges of Cu, Zn, Ni, Cr, Cd, Pb, and As (mg/kg) in roots were respectively 19.3-1440, 40.4-3120, 2.28-76.9, 22.6-250, 0.09-4.21, 84.1-1045, 5.32-157 (*pine*); 32.5-479, 60.2-1141, 3.03-53.3, 2.94-252, 0.09-1.84, 84.8-2380, 13.1-135 (*arbor*); 2.84-352, 7.25-721, 0.46-63.6, 2.95-410; 0.04-3.83, 15.2-2380, 3.17-16.1 (*fern*) (Fig. 3).

Generally, plants require appropriate amounts of trace elements to thrive. However, when in excess, these elements are toxic to plants. For instance, Zn and Cu are components of photosynthesis enzymes. They are generally required by plants at≤50 and 30 mg/kg, respectively. However, in this study, Zn and Cu concentrations in the root samples were 1.64 and 4.04 times those standards for normal plant growth [33].

Meanwhile, S8, located in the Dalingshan forest park, evidenced the highest total concentration of trace metals in the pine roots, primarily because of the high contents of Cu, Zn, Cd, Pb, and As. However, the high total concentrations of trace metals in arbor roots of S2 (Huangqishan Forest Park) and S8 (Dapingzhang

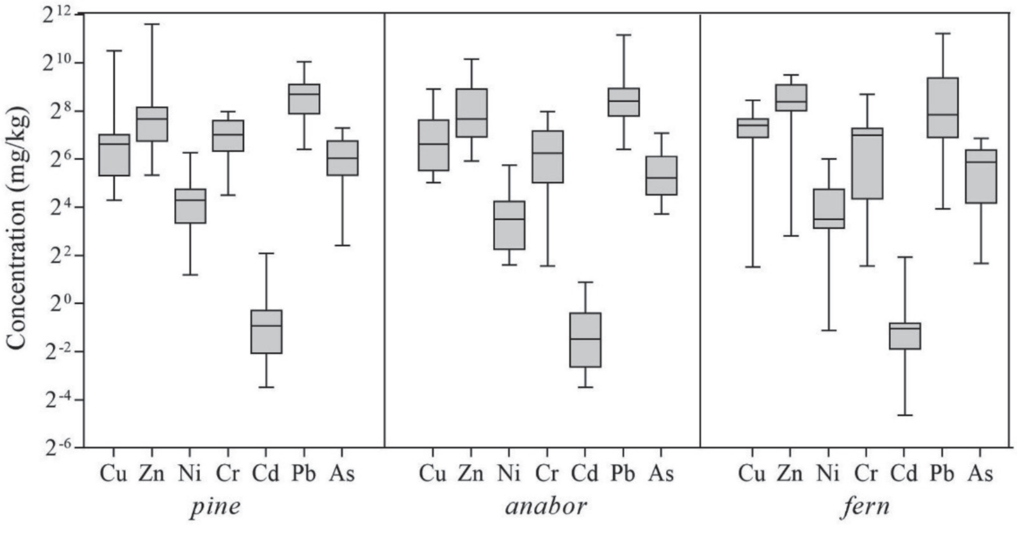

Fig. 3. Trace metal concentrations in plant roots (pines, arbors, ferns).

Forest Park) are attributed to Pb in S2, Cu, Zn, and Cd in S8. Furthermore, the sum trace metal concentration in the fern roots at S14 (in Yinpingshan Forest Park) was the highest (3260 mg/kg), dominated by As, Cu, and Pb. Thence, the parks and metals mentioned in this section require adequate environmental monitoring.

Characterization of Soil Pollution

The geological accumulation index (Igeo) is an indicator of trace metal pollution in soils by considering the metals' background values [25]. After calculating the Igeo of the trace metals in the six forest parks, we observed the following: the Igeo for Cu, Zn, Ni, Cr, and Pb belonged to class zero, i.e., unpolluted. Whereas, for As, Igeo<2, suggesting a fluctuation between unpolluted and moderately polluted soil. Cd evinced Igeo of 1.34-5.20, indicating moderate pollution to extreme pollution. The highest Igeo value of Cd in S4 soil (in Dapingzhang Forest Park) was 5.20 (Fig. 4a).

The Ef values for soil samples ranged from non-enriched to significant enrichment [25]. In the study, Mn was the chosen reference metal. The Ef ranges of Cr, Ni, and Zn were 0.42-4.62, 0.47-3.63, and 0.66-3.08, respectively, meaning depletion-to-minimum-to-moderate enrichment ensued. However, depletion-to-minimum-to-extremely high enrichment was noted for Cd and As, with corresponding Ef ranges of 9.94-129 and 1.65-15.9 (Fig. 4b). Approximately 53% of the soil samples showed excessively high Cd enrichment, while 60% showed significant As enrichment. High As level was consistently observed in the forest soil as previous studies showed that As pollution had intensified in Dongguan soil. As mainly originated from mining and smelting slags, industrial wastewater from metallurgy, chemical industry, pesticide, dyestuff and tannery, etc. Moreover, use of arsenic-containing pesticides and coal combustion could also resulted in the release of arsenic into the environment. Further, Cd concentration has increased in recent years, thereby becoming highly enriched in the soil [30]. The presence of Cd in phosphate fertilizers has been opined as responsible for its enrichment of Cd in the surface layer [34].

The pollution coefficients (Cf) of Ni and Cr in the six forests were <1, indicating mild Ni and Cr pollutions. The Cf values in Tongsha Ecological Park indicated moderate polluted by Zn and Pb (Cf 1-3) (Fig. 4c). However, the other five parks evidenced low pollution by Zn and Pb (Cf<1). All the forests were moderately polluted by As (Cf = 1.31-1.88). Except for the Shuilianshan forest park with mild Cu pollution, the other parks showed moderate Cu pollution. Cd pollution was too high (Cf = 4.98-19.7) in all the sites. So far, the Cf values are similar to those of Ef. Therefore, severe Cd pollution, and various degrees of As pollution, were mainly caused by human activities such as electroplating, producing of pigments, plastic stabilizers, nickel-cadmium batteries and electronics in the study area.

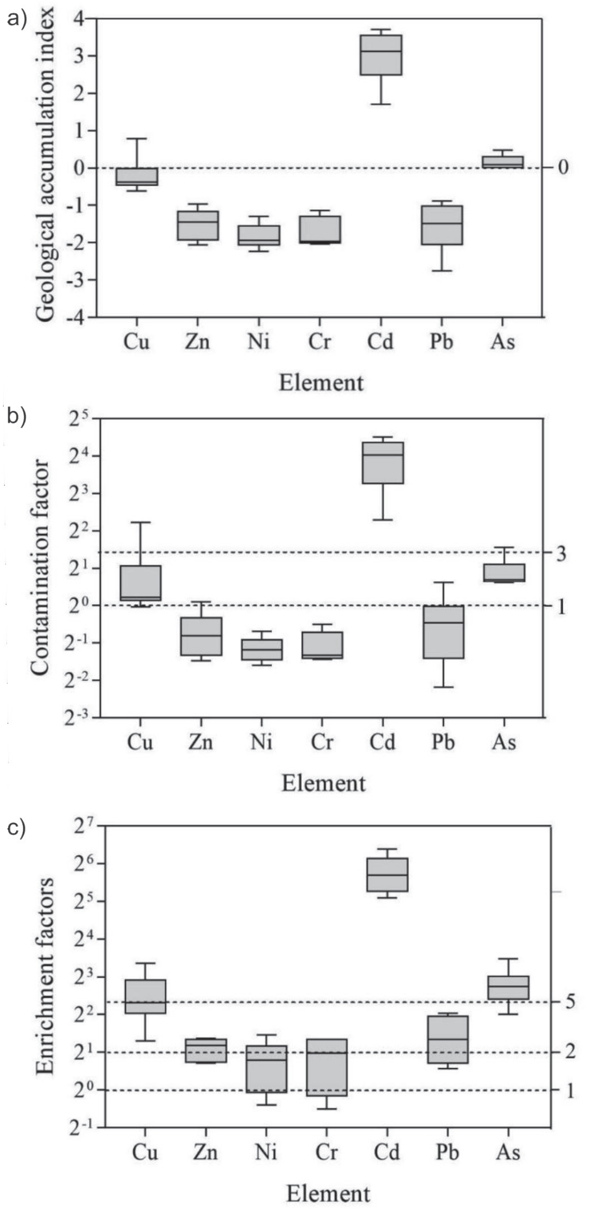

Fig. 4. The geological accumulation index, contamination factor and enrichment factor of trace metals in the surface soil. a) geological accumulation index and b) contamination factor; c) enrichment factor.

The multi-element pollution index is preferred to the single-element indexes, such as Ef and Cf [25, 34]. For example, the Modified Pollution Index (MPI) integrates the cumulative effects of multiple pollutants in the environment for assessment. The MPI values of the six forest parks ranged from 7.34 to 92.6, exhibiting heavy-to-severe pollution. Only a sampling site in Dapingzhang Forest Park with an MPI of 8.18 was not heavily polluted (Table S3). The highest MPI (92.6) was observed in Yinpingshan Forest Park, attributed to the elevated EF of Cd and As. Almost all the high MPI values could be ascribed to the high Ef values of Cd and As.

Modified degree of contamination (mCd), based on all the contamination factors (Cf), is used to analyze the degree of pollution [25, 35]. In this study, the mCd values of each forest park were derived from calculating the contamination degree of seven trace metals. The mCd of all the forest parks ranged from 1.15 to 11.01, suggesting unpolluted-to-severely-polluted degrees (Table S3). Specifically, for the Shuilianshan Forest Park, the mCd was <1.5, which indicates the park was unpolluted, whereas those of Huangqishan City Park and Dapingzhang Forest Park were moderately and severely polluted, respectively. Overall, S11 recorded the highest mCd, inferring Cd as the main pollutant. Further, we carried out the ecological risk assessment of the trace metals in Dongguan forest soil.

To comprehensively estimate the soils' trace metal pollution, we applied the Nemerow's pollution index (PI), which accounts for the pollution of all trace metals studied into account [28]. The PI values of the six forests, except that of S10, were within the range of 2.8-39.8. This range indicates that the study area was heavily polluted with toxic metals. Individually, the PI of Tongsha Ecological Park (18.3) and Dapingzhang Forest Park (39.8) were the highest. Furthermore, the higher Cf of Cu and Cd (in Dapingzhang Forest Park) and that of Cd (in Tongsha Ecological Park) led to the largest MPI in S11 and S4, respectively (Table S3).

Moreover, the BGI describes O horizon's adsorption capacity to pollutants, calculated based on the metal contents at various horizons [36]. The total BGI for the metals ranged from 0.16 to 9.98, demonstrating that O horizon's adsorption was low-to-moderate (Fig. S2). The most BGI values were <5, which proved that the adsorption of the trace metals was low-to-apparent. Overall, S11 (in Dapingzhang Forest Park) had high adsorption for Cu (BGI = 7.65) and Cd (BGI = 6.63), suggesting that Cu and Cd polluted the forest more than other metals. Furthermore, S4 (in Tongsha Ecological Park) had the highest BGI. Our results showed that the O horizon adsorbs Pb moderately, depending on the soil's physicochemical properties and metal concentrations.

Ecological Risk Assessment

Based on the soil background value in Guangdong, Er_i is calculated by the biotoxicity coefficient and ecological effects of trace metals [37]. The Er_i is determined for each trace metal toward evaluating its potential ecological risk. In general, the Er_i in the urban forest soils of Dongguan ranged from 0.31 to 1657. Generally, Zn, Ni, Cr, and Pb showed low ecological risk in all the sites ($Er_i<40$), with respective values of 0.31-1.81, 0.93-4.04, 0.51-1.79, and 1.02-14.31. However, Cu (Er_i = 57.89) and (As Er_i = 64.32) exhibited moderate ecological risk at S11 (in Dapingzhang Forest Park), while the Cu and As showed low ecological risk at other sampling sites. Additionally, Cd (Er_i = 114-1660) had high-to-very high risk in all the sites, indicating a severe potential ecological risk in Dongguan. Moreover, S11 (in the Dapingzhang Forest Park) evidenced the most serious ecological risk by Cu (Er_i = 57.6), As (Er_i = 64.3), and Cd (Er_i = 1660), 6.25, 3.39, and 3.64 times their respective averages (Table S4). Furthermore, the ecological risks of Cu, As, and Cd was related to the nearby electronic, plastic, and precision equipment industries.

Another hazard index of interest is the Hakanson potential ecological hazard index (RI), which evaluates trace metals' ecological risk in forest soils [38]. It integrates the potential ecological risks of multiple trace metals. The study area showed moderate-to-high ecological risk, with the RI value ranging from 139 to 1790. Due to the Er_i of Cu, the highest RI appeared in S11 (in Dapingzhang Forest Park). However, S4 (in Tongsha Ecological Park) and S8 (in Dalingshan Forest Park) also exhibited high ecological risk. The nearby industrial zone might contribute to their pollution of the Dalingshan Forest Park and Tongsha Ecological Park. The MRI values of all sampling sites were >300, ranging from 364 to 4160. These values indicate that the Dongguan urban forest soils are at ecological risk. Among the six forests, the Yinpingshan Forest Park showed high ecological risk in S12 (in Dongguan and Huizhou border, with some factories), having the highest MRI of 4160 (Table S5). Moreover, the Eri (114-1660) and MEri (299-3860) values for Cd in the sampling sites contributed the highest to the RI and MRI. In addition, S11, with a high MRI of 3670, also corroborated the RI deductions.

Sources Apportionment

Literature suggest that if a significant correlation exists among different trace metals, then they may possibly have the same source of contamination [39-40]. Pearson's correlation analysis (Table S6) indicated that no significant correlation exists between Cr and other metals except Ni. Zn and As, Ni and Cr, and Pb and As exhibited significant positive correlations ($p<0.05$). Yet observed were extremely strong positive correlations ($p<0.01$) between several pairs: Cu–Cd (0.864), Cu–As (0.955), Cd–As (0.911), Zn–Pb (0.829), Zn-Cd (0.702) and Cd-Pb (0.645). Therefore, we inferred that Cu, Zn, Cd, As, and Pb perhaps originated from the same source, while Cr and Ni might have come from another source.

Elsewhere, two principal components (PC) were extracted. The characteristic root values were >1, i.e., 3.94 and 1.66. The first two principal components reflected 79.91% of all the information of trace metal elements in the soils, indicating that the bulk of the data was obtained from the first two principal component analyses, whose proportions were 70.42% and 29.58%, respectively. The variation diagram in the rotated space is shown in Fig. S3. The 56.67% rate of variance for PC 1 was superior to PC 2 in analyzing the source of trace metals in Dongguan Forest soil. Cu, Zn, Cd, Pb, and As in PC 1 had higher positive load coefficients of 0.844, 0.843, 0.919, 0.789, and 0.899, respectively, whereas Ni and Cr in PC 2 evinced load factors of

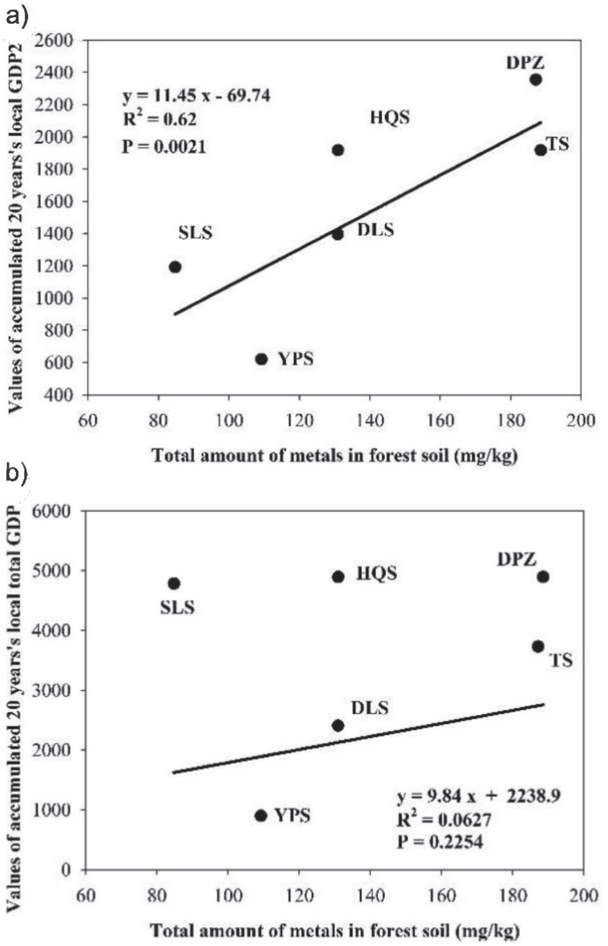

Fig. 5. Correlation of total metals in forest soil with cumulative twenty-year GDP. a) secondary GDP, b) total GDP.
GDP2: secondary GDP, GDP: total GDP.

0.755 and 0.875, respectively. By combining with the correlation analyses, Cu, Zn, Cd, Pb, and As might share the same source, while Ni and Cr might also be from the same source. These observations indicate that the trace metals in Dongguan urban forest soils mainly originated from two sources. Further, Cu, Zn, Pb, and As exhibited low potential ecological risks, while that of Cd was high, which might be from the nearby industrial activities such as electroplating, producing of pigments, plastic stabilizers, nickel-cadmium batteries and electronics, as well as agricultural use of metal-containing pesticides and insecticides. Besides, Cr and Ni showed low ecological risks, which perhaps came from natural sources.

Correlations of Metal Content with Accumulated Gross Domestic Product

The indices from geochemical accumulation and other pollution indices shown that the soils in forest were accumulated with a high amount of metal. The source apportionment suggested that the metals such as Cu, Zn, Pb, Cd, and As were mainly originated from human activities. Considering the rapid industrialization of the Dongguan area in the past 20 years, the relationship between the content of heavy metals in the soil and the regional GDP was analyzed. Results show that there is a significant correlation between the total amount of metals in forest soil and accumulated twenty years' gross domestic product for secondary industries for each local area ($p = 0.0021$) (Fig. 5a), while there is obvious correlation of metal content in soils with accumulated twenty years' total gross domestic product ($p = 0.2254$) (Fig. 5b). These results indicated that the industrial degree and development could be responsible for the content of all the metals in forest soils. To the best of our knowledge, this is the first time that a correlation between heavy metal contamination of forest soils and total industrial development has been reported.

Conclusions

We investigated the pollution characteristics of Zn, Cu, Cd, Cr, Ni, Pb, and As in the soils and roots of pines, arbors, and ferns in Dongguan's urban forests. We observed that Cd had gradually increased in recent years, whereas As and Cd had become the primary pollutants. The metals were mostly intercepted at the soil surface due to wet or dry deposition from atmospheric fallout. The plants in Dongguan forests were severely polluted with Pb, Cu, and As, with the ferns most metal-laden plant. Through the various pollution indexes, we observed that the urbanization gradients impact the Dongguan forest pollution. The concentrations and ecological risk assessment showed that Cd had a high potential risk on the Dongguan urban forest soils. Whereas, PCA indicated that Cr and Ni mainly originated from natural resources, while Cd, Pb, As, Cu, and Zn were from anthropogenic activities. Thence, the various forests require adequate monitoring of toxic metal concentrations, while an exigent approach to lower scenarios above permissible limits should be implemented immediately. Moreover, The level of total metals in soils from forest were significantly correlated with the accumulated twenty years' gross domestic product for secondary industries values and also the history of rapid industrialization.

Acknowledgments

The present study was supported by the Natural Science Foundation of Guangdong Province, China (2021B1515140023), Dongguan Science and Technology of Social Development Program (2020507140150, 20211800905532, 20211800904652) .

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.

Appendix A. Supplementary Data

The supplementary information and data associated with this article are shown in Figures S1–S3 and Tables S1–S5.

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Supplementary Materials

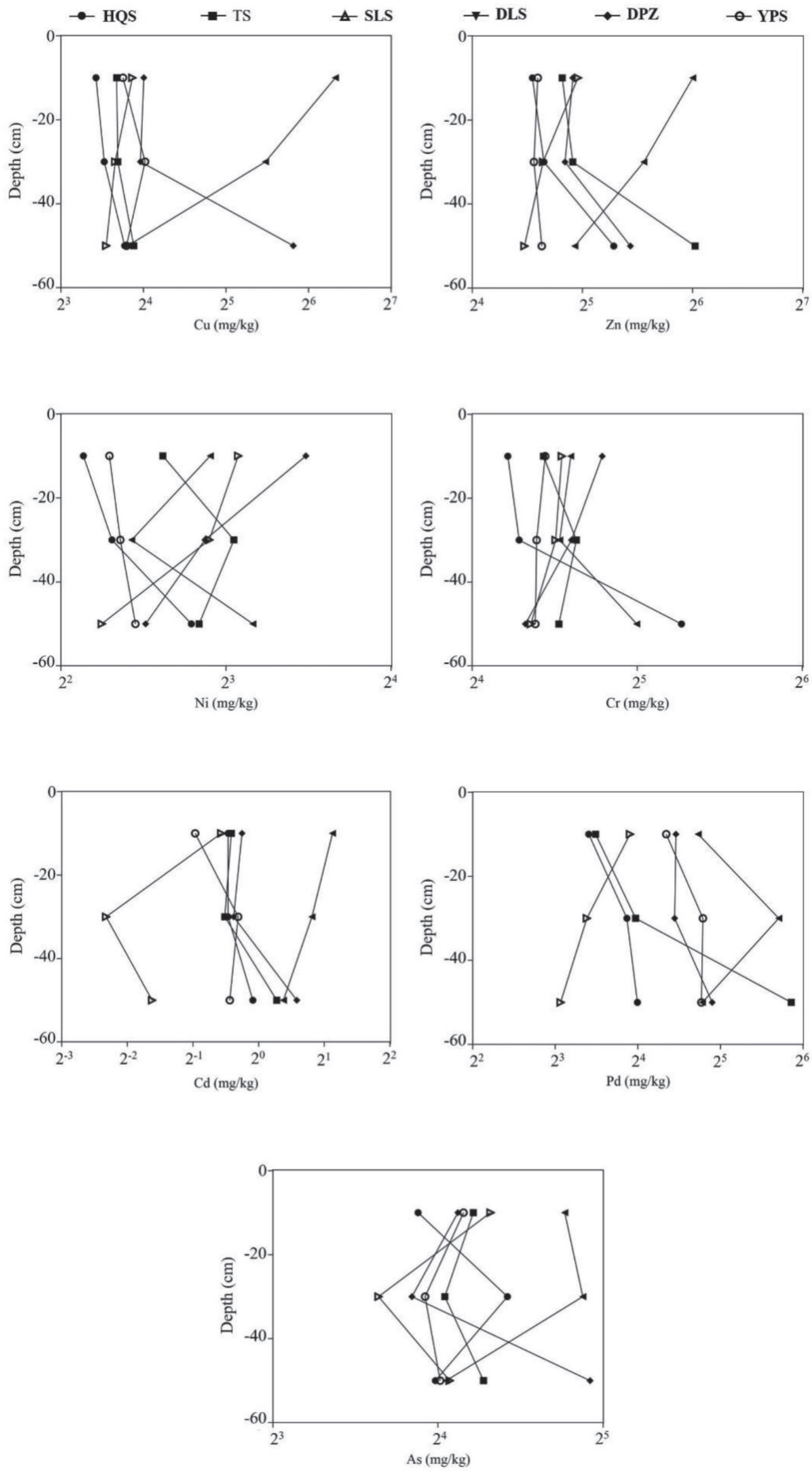

Fig. S1. Vertical distribution of Cu, Zn, Ni, Cr, Cd, Pb, and As in the soils.

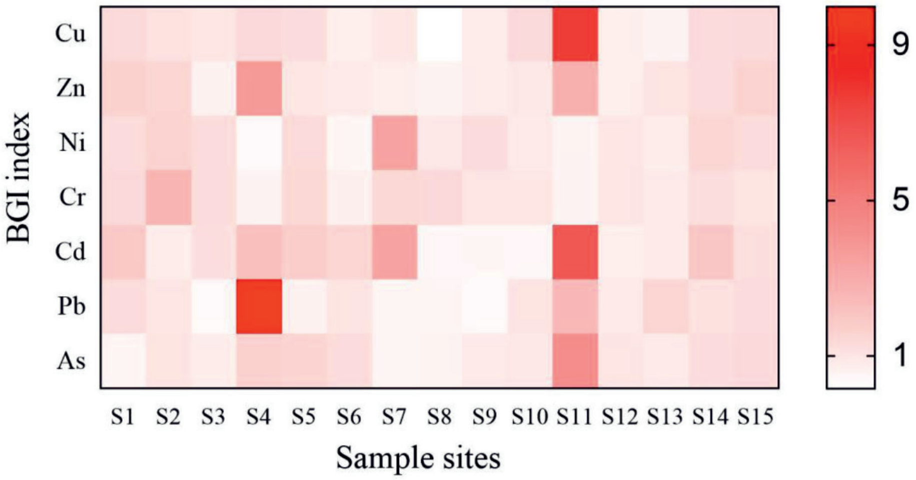

Fig. S2. BGI index of the trace metals in the soil samples.

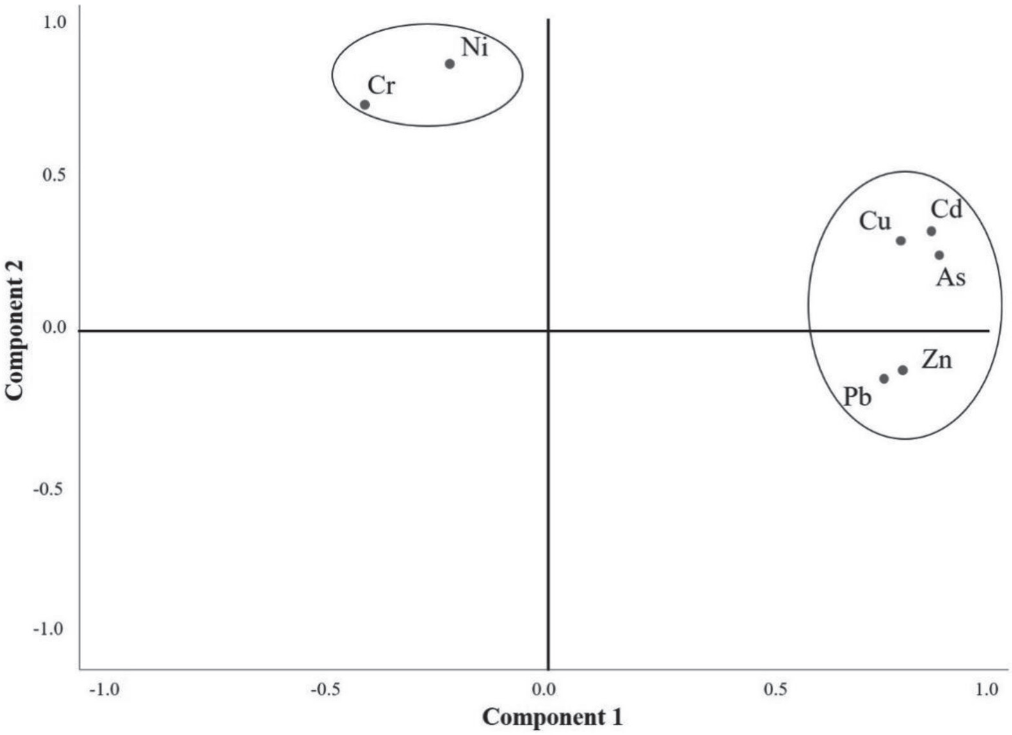

Fig. S3. The principal component (PC) analysis of trace metals in the surface soils of Dongguan forest.

Table S1. Indices in this study used to evaluate pollution of the study area.

Indices	Classification	Reference
$I_{geo} = \log_2(\frac{C_n}{1.5B_n})$	$I_{geo} < 0$: unpolluted $0 < I_{geo} < 1$: unpolluted to moderately polluted $1 < I_{geo} < 2$: moderately polluted $2 < I_{geo} < 3$: moderately to heavily polluted $3 < I_{geo} < 4$: heavily polluted $4 < I_{geo} < 5$: heavily to extremely polluted $I_{geo} > 5$: extremely polluted	Mazurek et al., 2016
$Ef = \frac{(metal/Fe)_{sample}}{(metal/Fe)_{UCC}}$	Ef < 2: depletion to minimum enrichment 2 ≤ Ef < 5: moderate enrichment 5 ≤ Ef < 20: significant enrichment 20 ≤ Ef < 40: very high enrichment Ef > 40: extremely high enrichment	Mazurek et al., 2016
$MPI = \sqrt{\frac{(Ef)^2_{\max} + (Ef)^2_{average}}{2}}$	MPI <1: Unpolluted 1 < MPI < 2: Slightly polluted 2 < MPI < 3: Moderately polluted 3 < MPI < 5: Moderately-heavily polluted 5 < MPI < 10: Heavily polluted MPI > 10: Severely y polluted	Brady et al., 2015
$C_f^i = \frac{C_s^i}{C_n^i}$	Cf < 1: low; 1≤ Cf < 3: moderate 3 ≤ Cf ≤ 6: considerable; Cf > 6: very high	Hakanson et al., 1980
$mC_d = \frac{\sum_{i=1}^{i=n} C_f^i}{n}$	mCd < 1.5: nil to very low 1.5 < mCd<2: low 2 < mCd < 4: moderate 4 < mCd < 8: high 8 < mCd < 16: very high 16 < mCd < 32: extremely high mCd > 32: ultra high	Abrahim and Parker, 2008
$PI = \sqrt{\frac{(Cf)^2_{\max} + (Cf)^2_{average}}{2}}$	PI < 0.7: Unpolluted 0.7 < PI < 1: Slightly polluted 1 < PI < 2: Moderately polluted 2 < PI < 3: Severely polluted PI > 3: Heavily polluted	Liu et al., 2015
$BAF = \frac{C_s}{C_n}$	--------	Li et al., 2010
$BGI = \frac{C_{nO}}{C_{nA}}$	BGI < 1: low; 1 < BGI <5: apparent; 5 < BGI < 10: moderate; 10 < BGI < 20: high BGI > 20: very high	Kowalska et al., 2018
$E_r^i = T_r^i \times C_f^i = T_r^i \times \frac{C_s^i}{C_n^i}$	$Er^i < 40$: low potential ecological risk; $40 \leq Er^i < 80$: moderate potential ecological risk; $80 \leq Er^i < 160$: considerable potential ecological risk; $160 \leq Er^i < 320$: high potential ecological risk; $Er^i \geq 320$: very high ecological risk	Hakanson et al., 1980
$RI = \sum_{i=1}^{n} E_i^i = \sum_{i=1}^{n} T_r^i \times C_f^i$	RI < 150: low ecological risk; 150 ≤ RI < 300: moderate ecological risk; 300 ≤ RI < 600: considerable ecological risk; RI ≥ 600: very high ecological risk.	Hakanson et al., 1980
$MRI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times E_f^i$	--------	Zhu et al., 2012

Where C_n represents the concentration of metal measured in soil samples; B_n represents the geochemical background value of the corresponding metal; $(metal/Fe)_{sample}$ represents the ratio of the metal concentration to the reference metal (Fe) concentration of the samples; $(metal/Fe)_{UCC}$ represents the ratio of the geochemical background value of the corresponding metal and the reference metal

(Fe); $(Ef)_{max}$, $(Ef)_{average}$, $(Cf)_{max}$ and $(Cf)_{average}$ represent the maximum of enrichment factor, average of enrichment factor, the maximum of contamination factor, average of contamination factor, respectively, C_s^i represents the concentration of a metal at a site and C_n^i is the geochemical background value of the corresponding metal, i = i^th^ element; C_{nO} and C_{nA} represent the concentration of metal in the O horizon and A horizon, respectively. T_r^i represents the toxic-response factor of trace metal and the T_r^i value for Cu, Zn, Ni, Cr, Cd, Pb, As were 5, 1, 5, 2, 30, 5 and 10, respectively. E_r^i represents the enrichment factor for each single element.

Table S2. Descriptive statistics for trace metals in roots of plants (pines, arbors, ferns) of the study area.

Species of plant		Cu	Zn	Ni	Cr	Cd	Pb	As
Pines (n = 15)	Minimum (mg/kg)	19.34	40.43	2.28	22.59	0.09	84.09	5.32
	Maximum (mg/kg)	1439.14	3122.93	76.94	250.09	4.21	1045.31	157.31
	Medium (mg/kg)	97.85	203.29	19.55	128.56	0.53	410.38	65.17
	Mean (mg/kg)	179.01	390.09	24.36	132.02	0.72	426.60	70.45
	CV (%)	1.97	1.95	0.79	0.48	1.40	0.63	0.65
Arbors (n = 15)	Minimum (mg/kg)	32.54	60.20	3.03	2.94	0.09	84.79	13.12
	Maximum (mg/kg)	479.41	1140.99	53.26	252.36	1.84	2257.77	135.33
	Medium (mg/kg)	98.74	203.52	11.29	75.72	0.36	341.51	37.26
	Mean (mg/kg)	132.25	296.75	15.04	97.18	0.55	486.38	52.46
	CV (%)	0.88	0.96	0.90	0.80	0.95	1.12	0.70
Ferns (n = 15)	Minimum (mg/kg)	2.84	7.25	0.46	2.95	0.04	15.21	3.17
	Maximum (mg/kg)	351.52	720.61	63.58	409.80	3.83	2378.77	116.06
	Medium (mg/kg)	169.93	333.10	11.36	126.65	0.49	230.56	58.71
	Mean (mg/kg)	152.98	352.79	18.43	128.46	0.69	518.08	57.66
	CV (%)	0.58	0.59	0.91	0.83	1.32	1.21	0.64

Table S3. The results of multi-element pollution index for the surface soil.

	PI	MPI	RI	MRI
HQS (n = 2)	10.27±4.44	47.03±46.1	455.56±184.26	2067.93±1993.54
TS (n = 2)	13.23±7.13	28.63±25.43	588.15±314.51	1271.63±1125.57
SLS (n = 2)	3.59±0.38	24.87±11.43	172.82±15.81	1192.80±534.09
DLS (n = 2)	14.17±1.83	60.31±1.98	622.04±78.61	2648.01±95.27
DPZ (n = 3)	16.39±20.37	36.49±39.5	741.29±912.94	1651.52±1766.23
YPS (n = 4)	8.08±3.98	38.15±38.1	364.03±175.27	1715.05±1704.63

All data are presented as Mean±SD.

*HQS: Huangqishan city park; TS: Tongsha ecological park; SLS: Shuilianshan forest park; DLS: Dapingzhang forest park; DPZ: Dalingshan forest park; YPS: Yinpingshan forest park

Table S4. The results of potential ecological risk assessment for the surface soil.

Sampling area*	Er (Cu)	Er (Zn)	Er (Ni)	Er (Cr)	Er (Cd)	Er (Pb)	Er (As)	RI	Ecological risk
HQS (n = 2)	5.7±0.38	0.64±0.02	2.4±0.14	1.41±0.54	427.9±187.3	2.13±0.04	15.38±2.79	455.56±184.26	Considerable
TS (n = 2)	6.16±0.98	1.07±1.05	2.48±2.2	0.84±0.47	551.05±297.19	7.72±9.31	18.82±8.64	588.15±314.51	Considerable
SLS (n = 2)	4.89±0.31	0.36±0.08	1.65±0.59	0.75±0.04	147.73±16.07	1.12±0.13	16.33±0.27	172.82±15.81	Moderate
DLS (n = 2)	5.85±1.38	0.5±0	3.11±0.95	1.17±0.02	591.65±77.24	3.67±1.07	16.09±4.8	622.04±78.61	very high
DPZ (n = 3)	23.5±29.82	0.71±0.53	1.98±0.32	0.73±0.04	680.96±848.59	3.98±4.1	29.42±30.24	741.29±912.94	very high
YPS (n = 4)	5.79±7.47	0.41±1.14	1.9±1.47	0.76±12.2	335.84±99.39	3.64±3.87	15.69±13.37	364.03±175.27	Considerable

All data are presented as Mean±SD.

*HQS: Huangqishan city park; TS: Tongsha ecological park; SLS: Shuilianshan forest park; DLS: Dapingzhang forest park; DPZ: Dalingshan forest park; YPS: Yinpingshan forest park. Er : ecological risk

Table S5. Correlation coefficient matrix of the trace metals in the soil samples.

	Cu	Zn	Ni	Cr	Cd	Pb	As
Cu	1.000	0.056	0.958	0.467	0.000	0.157	0.000
Zn	0.503	1.000	0.261	0.353	0.004	0.000	0.011
Ni	0.015	−0.310	1.000	0.015	0.729	0.437	0.770
Cr	−0.204	−0.258	0.612^{*}	1.000	0.649	0.196	0.435
Cd	0.864^{**}	0.702^{**}	0.098	−0.128	1.000	0.009	0.000
Pb	0.385	0.829^{**}	−0.217	−0.354	0.645^{**}	1.000	0.029
As	0.955^{**}	0.633^{*}	−0.082	−0.218	0.911^{**}	0.562^{*}	1.000

The left lower part is the correlation coefficient; the right upper part is the significance level

** indicates a significant correlation between the two elements ($p<0.01$, two-tailed)

* indicates a significant correlation between the two elements ($p<0.05$, two-tailed)

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