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Assessment of Soil Heavy Metal Pollution and Its Ecological Risk for City Parks, Vicinity of a Landfill, and an Industrial Area within Guangzhou, South China

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Abstract: As a primary sink of pollutants, urban soil heavy metal pollution and its influence on urban residents and ecosystems has been becoming one of the most important environmental problems. In the present study, four indices, the Geoaccumulation index (I_{geo}), improved Nemerow index (I_{MN}), degree of contamination (mC_d), and contamination security index (CSI), as well as potential ecological risk (RI), were used to evaluate individual or integrated heavy metal pollution and its ecological risk for soil samples collected from city parks, the vicinity of a landfill, and an industrial area within the city of Guangzhou. The results indicated that the improved Nemerow index (I_{MN}) calculated from the Geoaccumulation index was suitable for heavy metal pollution assessment of soils within landfills and industrial areas. As for soils collected from city parks, degree of contamination (mC_d) was more suitable than I_{MN}. Heavy metals Cd, Hg, Zn, and As were the main pollution elements in urban soils of Guangzhou. Potential ecological risks were mainly caused by Cd and Hg in urban soil of Guangzhou. Soil samples collected from city parks and the vicinity of the industrial area were moderately to highly and even extremely seriously polluted by heavy metals. Differing from the traditional cognition of the public, the ecological impact of heavy metal in soil in the vicinity of the landfill was similar to or even better than that within city parks.

Keywords: ecological risk; heavy metal pollution; surface soil; city parks; vicinity of landfill; industrial area; Guangzhou

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Citation: Zhou, H.; Ouyang, T.; Guo, Y.; Peng, S.; He, C.; Zhu, Z. Assessment of Soil Heavy Metal Pollution and Its Ecological Risk for City Parks, Vicinity of a Landfill, and an Industrial Area within Guangzhou, South China. *Appl. Sci.* **2022**, *12*, 9345. https://doi.org/10.3390/app12189345

Academic Editor: Anna Annibaldi

Received: 19 August 2022
Accepted: 15 September 2022
Published: 18 September 2022

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1. Introduction

With the rapid urbanization and industrialization during the last decades, urban environments have been experiencing serious deterioration. Consequently, health and wellbeing of urban residents are affected by this deteriorated environment. As an attribute of urban activities, urban surface soil is the primary sink of all pollutants, including heavy metals. Previous studies indicated that heavy metals within urban surface soil are harmful to public health and the urban ecosystem [1–4]. Excessive enrichment of heavy metals in urban soils can lead to the destruction of soil ecosystems and other environmental problems, and in more severe cases can threaten human health [5]. Regular exposure to individual heavy metal or mixtures of heavy metals may cause cancer, liver problems, and neurological, hematological, endocrine, reproductive, and other health system disorders [6–8]. Many previous studies indicated different characteristics of soil heavy metal pollution in different urban function areas [9–16]. Yan et al. [17] revealed that metal enrichment in roadside soils was affected by the level of urbanization and road age. Obvious soil heavy metal pollution also appeared at public playgrounds [18,19]. Heavy metal pollution of soils collected from urban parks [20–25] and the vicinity of landfills [26–28] attracted the most attention during the last years. Meanwhile, heavy metals within urban soils were proved to originate from industrial areas, landfills, and public activity areas [29–40]. However,

integration and comparison studies of soil heavy metal pollution for different function areas within a city are limited.

The study of soil heavy metal pollution for south China has been becoming a hotspot of environmental science due to the high geochemical background and the increasing human activities [41–51]. As the largest city in Southern China, many previous studies focused on heavy metal pollution of urban soil for different areas in Guangzhou. Compared to soil background values, heavy metals Cd, Cu, Hg, Mn, Pb, and Zn were significantly enriched in surface soil of Guangzhou due to a long history of urbanization and industrial activities [52–61]. However, few studies simultaneously concerned soil heavy metal pollution for different function areas in Guangzhou.

According to the conceptual FEP model proposed by Tatomir et al., the identification of pollution status which represents the "Feature" is an essential step for assessing the potential impact of heavy metal pollution [62]. As for the pollution assessment method, integrated pollution indices and the ecological risk index are useful tools that were widely used in previous studies. At least six indices were used for soil heavy metal pollution assessment by previous studies [21,63–67]. However, the assessed results using different methods were much different [11,14,43,55,68–75]. Therefore, selecting a proper index is a key for accurately acquiring the degree of contamination.

In the present study, heavy metal concentrations in surface soil samples collected from city parks, the vicinity of a landfill, and an industrial area within Guangzhou were simultaneously measured. Four pollution indices (I_{geo}, I_{MN}, mC_d, and CSI) and the potential ecological risk index (RI) were used to assess heavy metal pollution degree and its ecosystem risks, respectively. The main objective of this work is to enhance our understanding of the status of soil heavy metal pollution in Guangzhou, to reveal the differences in soil heavy metal pollution status and potential ecological risks for different urban function regions such as city parks, the vicinity of a landfill, and an industrial area, and to recommend more suitable assessment methods for evaluation of urban soil heavy metal pollution.

2. Materials and Methods

2.1. Study Area and Sampling

The study area, the city of Guangzhou, is the capital of Guangdong Province, located in South China (Figure 1). A subtropical monsoon climate characterized by being hot and humid with a mean annual temperature of 22 °C and mean annual rainfall of 1647 mm predominates the study area. Following to the guidelines of the Specification of Regional Ecogeochemistry Assessment and the Determination of Local Ecogeochemistry Assessment by the Ministry of Natural Resources of the People's Republic of China, two hundred and twenty-nine surface soil (0–20 cm) samples were collected from city parks, landfills, and industrial areas within Guangzhou's built-up area in 2015.

According to the distribution and developing history of city parks within Guangzhou, one hundred and thirty-two topsoil samples were collected from eleven parks within the city's built-up area. Forty-one topsoil samples were collected from the vicinity of the Likeng landfill, which was put into use in 1992 and closed in April 2004. The Huangpu industrial zone with a state-owned large-scale enterprise Huangpu power plant at the southeast, a large oil storage of petrochemical plants at the northeast, and some other industries such as metal processing plants, concrete mixing plants, and fly ash plants within the area, was selected for the representative industrial area. Fifty-six topsoil samples were collected from the Huangpu industrial zone. The distribution of sampling sites is illustrated in the enlarged maps of Figure 1 for the three mentioned functional areas.

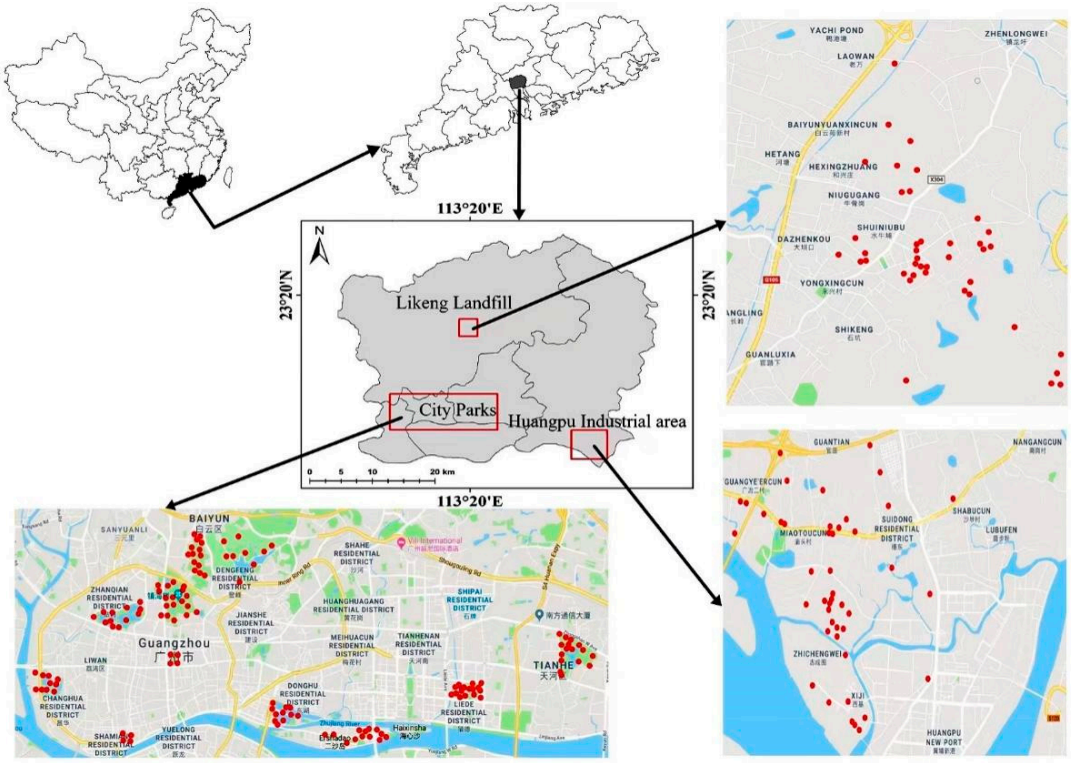


Figure 1. Location of the study area and distribution of sampling sites. Red dots represent sampling sites.

2.2. Heavy Metal Content Measurements

All samples were air dried at room temperature after sampling. After removing obvious gravel and roots, the samples were ground into a powder by sieving through a 74 μm mesh for element content measurements.

Heavy metal content measurements were performed at the ALS Minerals-ALS Chemex Laboratory, Guangzhou. An aliquot of a dry burning sample without organic matter was dissolved in aqua regia. Contents of heavy metals Copper (Cu), Nickel (Ni), and Cadmium (Cd) were determined using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700x). Zinc (Zn), Lead (Pb), and Chromium (Cr) contents were measured by an X-ray fluorescence spectrometry. Arsenic (As) and Mercury (Hg) contents were analyzed using atomic fluorescence spectrometry. The detection limits were the following (as mg/kg): 0.2 Cu, 0.2 Ni, 0.001 Cd, 2.0 Zn, 0.5 Pb, 1.0 Cr, 0.2 As, and 0.004 Hg. During the measurements, quality controlling samples including standard samples, duplicate samples, and blank samples were added among the detected samples according to the regulations stated in ISO17025.

2.3. Pollution Indices

Pollution indices are widely used for comprehensive assessment of soil pollution degree [21,76–79]. Four pollution indices, Geoaccumulation index (I_{geo}), improved Nemerow index (I_{MN}), degree of contamination (mC_d), and contamination security index (CSI) were calculated for each sample to assess their heavy metal pollution degree. The calculating formulas and the pollution classification criteria used for each index are listed in Table 1. In order to reflect the differentiation of the specific area, the average values of A horizons in Guangdong Province (167 typical profiles, 33 main profiles) were used as the local natural background values.

Table 1. Indices of pollution used in this study.

Index	Description and Aim of Use	Formula	Classifications of Contamination Situation
Geoaccumulation index (I_{geo})	I_{geo} determines metal contamination in soil with respect to natural background level as a reference [80]. Classification criteria according to Förstner et al. [81].	$I_{geo} = log_2\left[\frac{C}{k*Bn}\right]$ C—measured heavy metal content, B_n—background value of heavy metal content, k—the petrogenesis effect, usually set to 1.5 [82,83]	≤0: unpolluted; 0–1: slightly polluted; 1–2: moderately polluted; 2–3: moderately to highly polluted; 3–4: highly polluted; 4–5: seriously polluted; ≥5: extremely seriously polluted.
Improved Nemerow index (I_{MN})	I_{MN} allows the assessment of the overall degree of soil pollution and includes the contents of all analyzed elements. The contamination factor is substituted by the I_{geo} [76,77].	$I_{MN} = \sqrt{\frac{\left({I_{geo_{max}}}^2 + {I_{geo_{ave}}}^2\right)}{2}}$ $I_{geo_{max}}$—the maximum value of the I_{geo} of all metals in a sample, $I_{geo_{ave}}$—the arithmetic mean of the I_{geo} of all metals in a sample	<0.5: uncontaminated; 0.5–1: slightly contaminated; 1–2: moderately contaminated; 2–3: moderately to heavily contaminated; 3–4: heavily contaminated; 4–5: heavily to extremely contaminated; ≥5: extremely contaminated.
Degree of contamination (mC_d)	mC_d assesses the degree of contamination in soil [84,85].	$mC_d = \frac{\sum_{i=1}^{n} \frac{C_s^i}{C_n^i}}{n}$ C_s^i—measured heavy metal content, C_n^i—background value of heavy metal content	<1.5: nil to very low degree of contamination; 1.5–2: low degree of contamination; 2–4: moderate degree of contamination; 4–8: high degree of contamination; 8–16: very high degree of contamination; 16–32: extremely high degree of contamination; ≥32: ultra-high degree of contamination.
Contamination security index (CSI)	*CSI*, introduced by Pejman et al. [78], is informative about the heavy metal pollution intensity in soil.	$CSI = \sum_{i=1}^{n} w_i \left[\left(\frac{C}{ERL}\right)^{\frac{1}{2}} + \left(\frac{C}{ERM}\right)^2\right]$ w_i—weight for each heavy metal, calculated according to Pejman et al. [78], C—measured heavy metal content, effects range low (ERL), and effects range median (ERM), given by Long et al. [86]	<0.5: uncontaminated; 0.5–1.5: low severity of contamination; 1.5–2.5: moderate severity of contamination; 2.5–3: moderate to high severity of contamination; 3–4: high severity of contamination; 4–5: very high severity of contamination; ≥5: ultra-high severity of contamination.

2.4. Potential Ecological Risk Index

Potential ecological risk index (*RI*), introduced by Hakanson [84], is an indicator to assess degree of environmental risk caused by heavy metals in water and air, as well as in soil. *RI* is calculated for each sample according to the following equation.

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \frac{C^i}{C_n^i}$$

where E_r^i is the potential ecological hazard coefficient of heavy metal i, T_r^i is the toxicity coefficient of heavy metal i, C^i is the measured heavy metal content, and C_n^i is the background value of heavy metal content.

As suggested by Hakanson [84] and revised by Xu et al. [87], the values of T_r^i used in the present study are Zn = 1 < Cr = 2 < Pb = Cu = Ni = 5< As = 10 < Cd = 30 < Hg = 40. Again, the average values of layer A soils in Guangdong Province are used for C_n^i. Combined considering several references [78,84], the classification criteria of E_r^i and *RI* in this study are listed in Table 2.

Table 2. Classification criteria of potential ecological risk index.

E_r^i Values	*RI* Values	Risk Intensity
<40	<150	Low ecological risk
40–80	150–300	Moderate ecological risk
80–160	300–600	Considerable ecological risk
160–320	600–1200	High ecological risk
≥320	≥1200	Very high ecological risk

3. Results

3.1. Heavy Metal Concentrations

A statistical summary of heavy metal content as well as the background value of surface soil (A horizon) in Guangdong is listed in Table 3.

Table 3. Statistical summary of heavy metal contents (unit: mg/kg).

Area		Cu	Zn	Ni	Pb	Cr	As	Cd	Hg
City parks	Min	8.2	18.0	6.4	22.2	13.0	6.3	0.012	0.006
	Max	240.0	3660.0	65.1	564.0	145.0	305.0	15.800	11.950
	Ave	44.0	203.0	19.9	105.0	51.7	34.7	0.599	0.804
	Std.	32.0	351.7	9.2	77.1	18.1	35.7	1.502	1.329
	CV (%)	73.0	173.2	46.4	73.4	35.0	103.2	251.8	165.4
Landfill	Min	8.3	13.0	4.8	14.8	12.0	5.3	0.010	0.006
	Max	160.0	268.0	127.5	5420.0	145.0	604.0	3.150	0.217
	Ave	24.4	83.2	19.3	177.7	44.6	56.4	0.310	0.105
	Std.	28.2	55.4	22.8	839.6	27.6	109.2	0.515	0.059
	CV (%)	115.6	66.6	118.4	472.5	61.9	193.8	165.8	59.2
Industrial area	Min	9.6	56.0	8.0	46.0	21.0	5.1	0.101	0.043
	Max	2365.0	5640.0	136.8	942.0	1120.0	78.5	17.650	1.570
	Ave	170.4	601.1	30.8	204.6	107.0	19.3	1.415	0.353
	Std.	407.2	1080.6	28.9	200.4	162.2	14.0	2.452	0.312
	CV (%)	239.0	179.8	93.7	97.9	151.6	72.5	173.3	88.5
Background value of surface soil (A horizon) in Guangdong) *		17.0	47.3	14.4	36.0	50.5	8.9	0.056	0.078

* Data taken from The Background Values of Soil Elements in China [88].

Except the Cr content within surface soil around the landfill, average contents of all heavy metals within surface soil exceeded their background values. Even the minimum values of Zn, Pb, and Cd contents within surface soil exceeded their background values for the industrial area. According to the average contents listed in Table 3, contents of Cu, Zn, Ni, Cr, and Cd within surface soil increased from landfill, to city parks, to industrial area. Surface soil Pb content increased from city parks, to landfill, to industrial area. Surface soil As content increased from industrial area, to city parks, to landfill. Surface soil Hg content increased from landfill, to industrial area, to city parks.

3.2. Pollution Indices

The evaluated results of pollution indices listed in Table 1 are illustrated in Figure 2 for city parks, landfill, and industrial area.

The results of the Geoaccumulation index and I_{MN} revealed that more than 70% of the measured 132 samples collected from city parks were moderately and moderately to highly polluted by heavy metals (Figure 2a). Samples showing slight or high to serious heavy metal pollution were about 10% of the total 132 samples. According to the results of the Geoaccumulation index for each heavy metal element illustrated in Figure 2a, almost all samples were unpolluted or slightly polluted by Ni and Cr. Most samples were slightly or moderately polluted by Cu, Zn, Pb, and As. However, most soil samples collected from city parks were moderately, moderately to highly, highly, seriously, or even extremely seriously polluted by Cd and Hg. Therefore, it can be concluded that the main pollution elements in urban city park soils in Guangzhou were Cd and Hg.

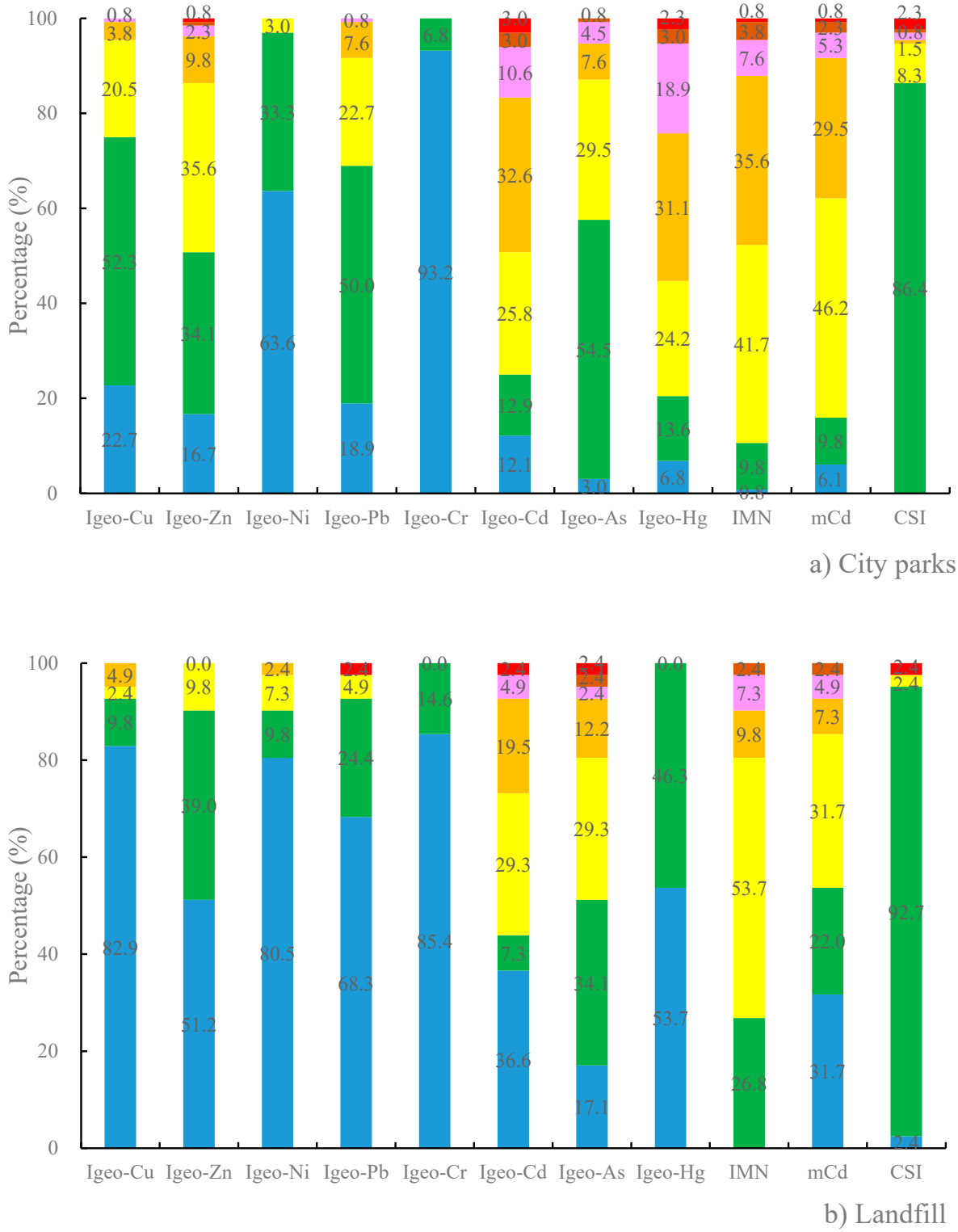


Figure 2. *Cont*.

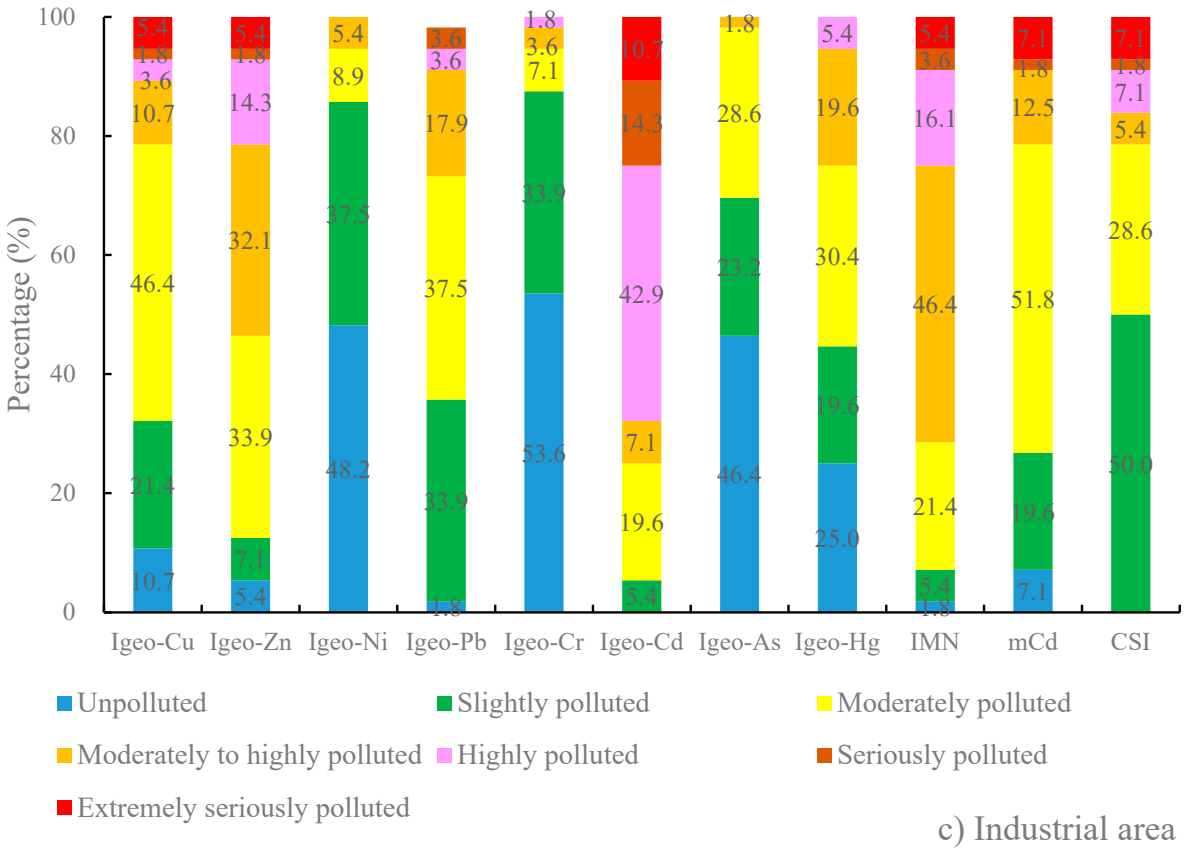


Figure 2. Percentage composition of different pollution degree based on calculation results of I_{geo} for every element and I_{MN}, mC_d, and *CSI* for eight heavy metals (**a**) city parks, (**b**) landfill, and (**c**) industrial area.

As for soil samples collected from the vicinity of the landfill, 73.2% of all samples were moderately to seriously polluted by heavy metals. All samples were clean or were slightly polluted by heavy metals Cr and Hg. More than ninety percent of the measured samples were unpolluted or slightly polluted by heavy metals Cu, Zn, Ni, and Pb. However, 56.1% and 48.8% of the measured samples collected from the vicinity of the landfill were moderately, moderately to highly, highly, and seriously polluted by Cd and As, respectively (Figure 2b).

As for the calculated results of I_{MN} for soil samples collected from the industrial area, nearly 90% samples were moderately to seriously polluted by heavy metals. Furthermore, some samples collected from the industrial area were even extremely seriously polluted by HMs (Figure 2c). Fortunately, most surface soil samples collected from the industrial area showed no or only slight pollution by Ni (85.7%), Cr (87.5%), and As (69.6%). Of the measured soil samples collected from the industrial area, 32.1%, 35.7%, and 44.6% were unpolluted or slightly polluted by heavy metals Cu, Pb, and Hg. For all measured soil samples from the industrial area, percentages of moderate Cu, Pb, and Hg pollution were 46.4%, 37.5%, and 30.4%, respectively. More than half of the measured soil samples collected from the industrial area were moderately to highly, highly, seriously, and extremely seriously polluted by Zn. Concerning Cd in soil within the industrial area, all measured samples were polluted at different degrees. The percentages of measured samples under slight, moderate, moderate to high, high, serious, and extreme serious pollution degree were 5.4%, 19.6%, 7.1%, 42.9%, 14.3%, and 10.7%, respectively (Figure 2c). Therefore, it can be concluded that Cd and Zn are the main heavy metal pollution elements in surface soils of the industrial area in Guangzhou.

4. Discussion

4.1. Comparison of Assessment Results Using Different Indices

Comparing the pollution evaluation results illustrated in Figure 2, it can be obviously seen that soil heavy metal pollution in both the comprehensive results and each element descended from the industrial area, to city parks, to the vicinity of the landfill. Except Cr and Ni in city park soils and Hg and Cr in soils in the vicinity of the landfill, other heavy metals appeared with different enrichment degrees in surface soils of different function areas within Guangzhou (Figure 2). Furthermore, the pollution degree of each element in soils of city parks, the vicinity of the landfill, and the industrial area were decreased following the order Cd, Hg, Zn, As, Pb, and Cu; Cd, As, Zn, Ni, Pb, and Cu; and Cd, Zn, Cu, Pb, Hg, As, Ni, and Cr, respectively. Despite Cd being the most important heavy metal pollutant in soils collected from the three function areas, the percentages of different Cd pollution degrees were much different (Figure 2). Heavy metals Hg, As, and Zn were also important heavy metal pollutants for soils of city parks, the vicinity of the landfill, and the industrial area, respectively.

A suitable method for analysis is the premise of regional soil pollution assessment. Compared the results of I_{MN}, mC_d, and *CSI* illustrated in Figure 2, it can be seen that comprehensive pollution degrees classified by I_{MN} and mC_d were similar for soil samples collected from city parks. However, pollution degrees classified by *CSI* were much weaker than those classified by I_{MN} and mC_d for the three function areas, indicating that heavy metal pollution assessed by *CSI* may be underestimated for urban surface soils. Results of I_{MN} indicated that percentages of measured soil samples under clean and slight pollution were 26.8% and 7.2% for samples collected from the vicinity of the landfill and the industrial area, respectively. Meanwhile, these percentages classified by calculated results of mC_d were 53.7% and 26.7% for the vicinity of the landfill and the industrial area, respectively (Figure 2b,c). Since obvious significant differences appeared between assessing the results of I_{MN} and mC_d (Figure 2), comprehensive assessment results by previous studies are summarized in Figure 3 to compare with the calculated results of I_{MN} and mC_d in the present study.

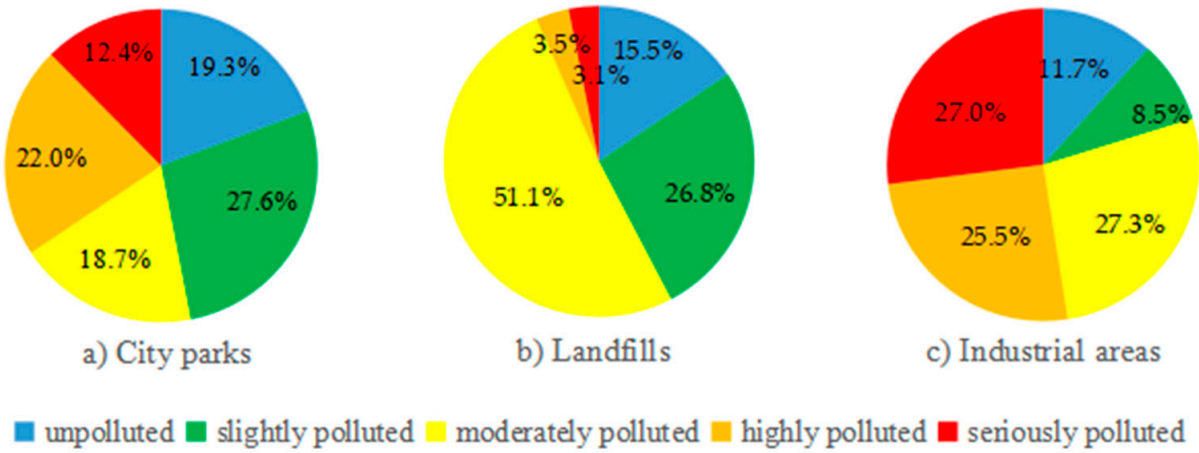


Figure 3. Soil heavy metal pollution assessment results by previous studies for the city of Guangzhou. (**a**) City parks (revised from [13,51,56,67,72]), (**b**) Landfills (revised from [14,55,59,73]), and (**c**) Industrial areas (revised from [13,14,47,49,53,57,60,61]).

As illustrated in Figure 3a, previous studies indicated that no more than 35% (about 34.4%) of measured soil samples suffered high or serious pollution of heavy metals. The calculated results of mC_d illustrated in Figure 2a are similar to the mentioned results of previous studies. However, the calculated I_{MN} results of the present study illustrated in Figure 2a indicated nearly half of the measured samples (about 47.7%) were under high or serious pollution. These results indicated that heavy metal pollution might be overesti-

mated for soils collected from city parks using I_{MN}. As for soils collected from the vicinity of landfills, the percentage of slight and moderate pollution assessed by calculated results of I_{MN} was almost consistent with these results of previous studies (Figures 2b and 3b). Meanwhile, the percentages of unpolluted and moderately polluted samples assessed by the calculated mC_d were twice as much as and half of the results of previous studies, respectively (Figures 2b and 3b). From Figures 2c and 3c, the percentages of seriously polluted samples assessed by results of I_{MN} and mC_d for soils collected from the industrial area were consistent with and much lower than the results reported in previous studies, respectively. The percentages of unpolluted and slightly polluted soil samples within the industrial area assessed by the I_{MN} results were much lower than the results of previous studies (Figures 2c and 3c). Furthermore, the percentages of highly polluted soil samples assessed by the calculated I_{MN} and mC_d results were much higher and lower than the results of previous studies, respectively (Figures 2c and 3c). Since heavy metals are continuously accumulated in urban soils, it is reasonable that the pollution degree of the present study is a little bit more serious than previous studies. Therefore, compared to the results of I_{MN}, the pollution situation may be underestimated by the results of mC_d. Thus, it can be concluded that I_{MN} is more suitable for heavy metal pollution assessment for soils within the vicinity of landfills and industrial areas of Guangzhou.

4.2. Ecological Impact of Heavy Metal Pollution

The calculated results of potential ecological risk for every element (E) as well as total potential ecological risks (*RI*) of the measured eight heavy metals are illustrated in Figure 4 for city parks, landfill, and industrial area.

Although some samples were under different degrees of pollution, potential ecological risks caused by heavy metals Cu, Zn, Ni, Pb, Cr, and As can be ignored for all surface soil samples collected from the three different function areas. However, surface soil samples collected from the city parks, landfill, and industrial area are suffering some ecological risks from Cd and Hg (Figure 4). Of the measured samples collected from city parks and the industrial area, 54.5% and 80.4% were subjected to high or very high ecological risks caused by Cd, respectively. The percentages of measured samples under high and very high ecological risks of Hg were 72.7% and 46.4% for city parks and the industrial area, respectively (Figure 4a,c). These results are consistent with previous studies on the ecological risk of soil heavy metals in urban parks, landfills, industrial areas, and urban areas in Guangzhou and urban soils in China [48,53,57,59,61,89]. Meanwhile, only 26.8% and none of the measured soil samples collected from the landfill vicinity were subjected to high or very high ecological risks caused by Cd and Hg, respectively. Totally, the percentages of measured soil samples collected from city parks, the vicinity of the landfill, and the industrial area under high or very high ecological risks caused by soil heavy metals were 44.7%, 12.2%, and 71.5%, respectively (Figure 4). These results indicated that the soil environment within the vicinity of the landfill was better than that within city parks, which was much different from the public's traditional cognition.

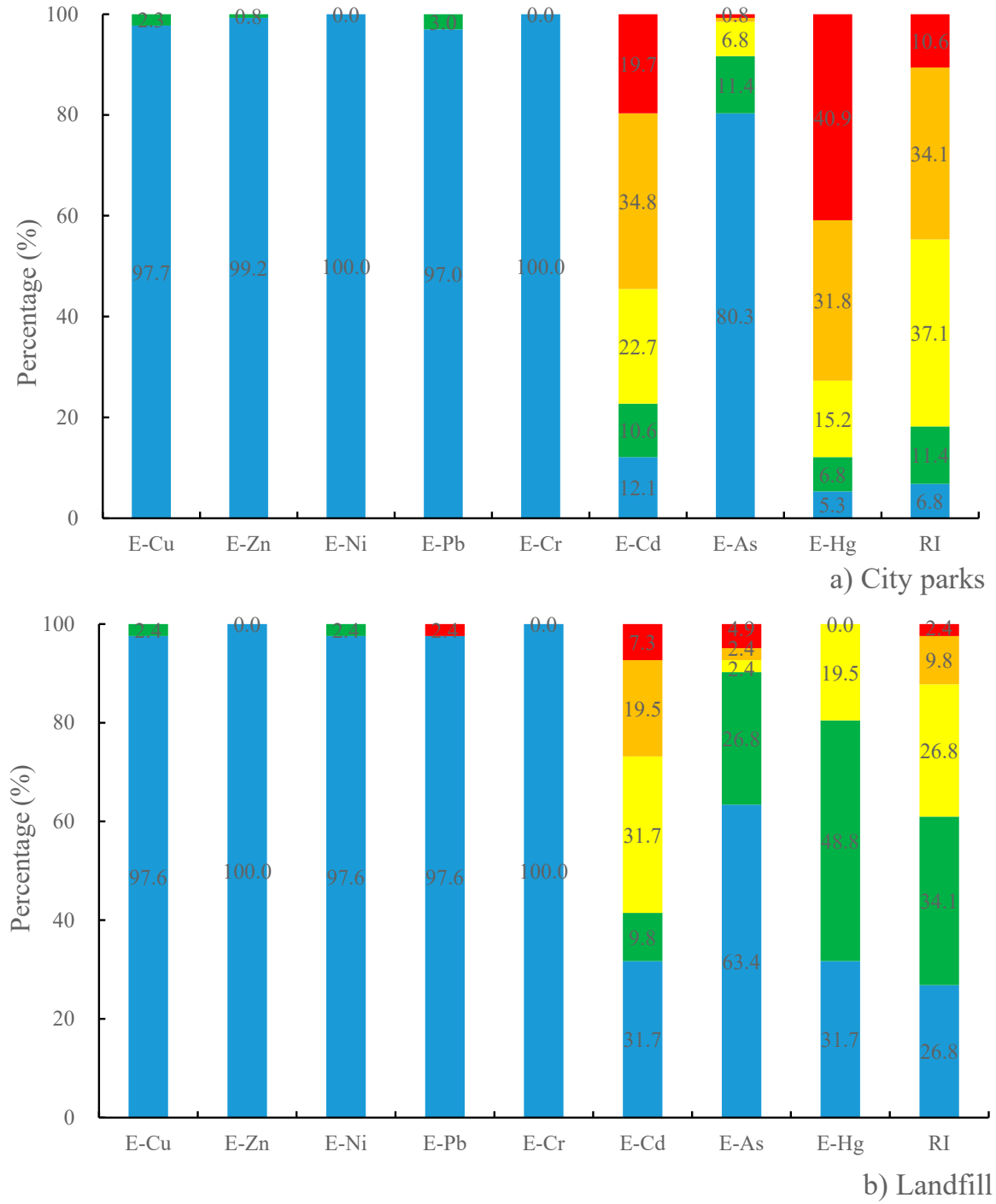


Figure 4. *Cont.*

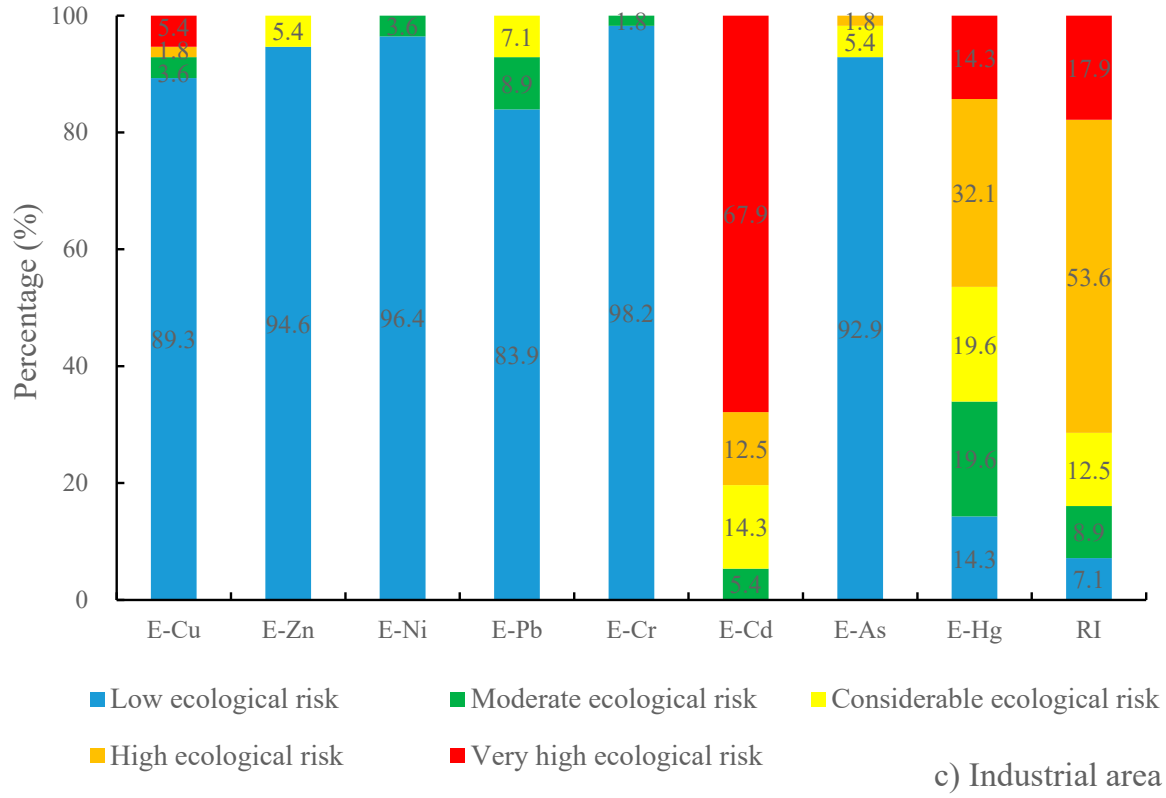


Figure 4. Percentage composition of different potential ecological risks caused by soil heavy metal based on calculation results of E for every element and RI for eight heavy metals: (**a**) city parks, (**b**) landfill, and (**c**) industrial area.

5. Conclusions

The identification of pollution status is an essential step for assessing the potential impact of urban soil heavy metal pollution. However, assessed results were much different using different methods. Therefore, selecting a suitable method should be one of the most important aspects of heavy metal evaluation. Based on the mentioned results of the discussion, the following conclusions that can be used for further relevant research are acquired.

The improved Nemerow index (I_{MN}) calculated based on the Geoaccumulation index is more suitable for heavy metal pollution assessment for soils within the vicinity of landfills and industrial areas of urban areas. Meanwhile, the degree of contamination (mC_d) is more suitable than I_{MN} for soil heavy metal pollution assessment for city parks.

The pollution degree of each element in soils of city parks, the vicinity of the landfill, and the industrial area were decreased following the order: Cd > Hg > Zn > As > Pb > Cu; Cd > As > Zn > Ni > Pb > Cu; and Cd > Zn > Cu > Pb > Hg > As > Ni > Cr, respectively. Heavy metals Cd, Hg, Zn, and As were the main pollution elements in urban soils of Guangzhou.

Considerable, high, to very high ecological risk was caused by heavy metal pollution of most samples collected from city parks and the industrial area. Meanwhile, low to moderate ecological risk was caused by heavy metal pollution of most samples collected from the vicinity of the landfill. Soil heavy metal potential ecological risks were mainly caused by Cd and Hg for the investigated three function areas. That the soil environment within the vicinity of the landfill was better than that within city parks differed from the public's traditional cognition.

Author Contributions: Conceptualization, T.O. and Y.G.; methodology, H.Z. and C.H.; investigation, Y.G., S.P. and Z.Z.; data curation, T.O. and Y.G.; writing—original draft preparation, H.Z. and T.O.; writing—review and editing, H.Z. and T.O.; supervision, Z.Z.; project administration, Z.Z. and S.P.; funding acquisition, T.O. and Y.G. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Natural Science Foundation of China (Grant No. 41977261) and the Guangzhou Land Resources and Planning Commission (Grant No. GSZB22C0025G).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All the data analyzed/generated are included and available in the manuscript.

Acknowledgments: The authors are grateful to the anonymous reviewers for their constructive comments.

Conflicts of Interest: The authors declare no conflict of interest.

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