An Integrated H-G Scheme Identifying Areas for Soil Remediation and Primary Heavy Metal Contributors: A Risk Perspective

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Traditional sampling for soil pollution evaluation is cost intensive and has limited representativeness. Therefore, developing methods that can accurately and rapidly identify at-risk areas and the contributing pollutants is imperative for soil remediation. In this study, we propose an innovative integrated H-G scheme combining human health risk assessment and geographical detector methods that was based on geographical information system technology and validated its feasibility in a renewable resource industrial park in mainland China. With a discrete site investigation of cadmium (Cd), arsenic (As), copper (Cu), mercury (Hg) and zinc (Zn) concentrations, the continuous surfaces of carcinogenic risk and non-carcinogenic risk caused by these heavy metals were estimated and mapped. Source apportionment analysis using geographical detector methods further revealed that these risks were primarily attributed to As, according to the power of the determinant and its associated synergic actions with other heavy metals. Concentrations of critical As and Cd, and the associated exposed CRs are closed to the safe thresholds after remediating the risk areas identified by the integrated H-G scheme. Therefore, the integrated H-G scheme provides an effective approach to support decision-making for regional contaminated soil remediation at fine spatial resolution with limited sampling data over a large geographical extent.

Soils are being increasingly polluted as a result of growing urbanization, deforestation and industrialization. The wide spread and hazards of soil pollution are detrimental for both the environment and human beings1–3. Among soil pollutants, heavy metals are extremely hazardous due to their non-degradability, leaching ability, and massive accumulation4–6. With the boost of urbanization and industrialization, China has become the world’s leading heavy metal producer, resulting in the contamination of soils with high concentrations of heavy metals. These contaminated soils pose serious threats to human health and social stability7, 8.

To mitigate the harmful effects of heavy metal pollution, the Chinese government has announced the National Remediation Project of Heavy Metal Contaminated Soil (NRP-HMCS) across the country. However, its effects are greatly reduced due to inadequate financial support and inadequate recognition of areas with heavy metal pollution in need of remediation9,10. Soil quality standards have been employed in China since 1995 with the release of "Environmental Quality Standard for Soils GB 15618-95" to assess soil pollution. However, areas with concentrations exceeding the standard do not necessarily pose a serious health risk caused by polluted soil because of spatially differentiated population distribution and exposure pathways. In other words, heavily polluted areas

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As, Cu, Hg, and Zn range from 1.0 to 7.1 mg/kg, 2.5 to 40.4 mg/kg, 0.3 to 86.1 mg/kg, 0.2 to 1.5 mg/kg, and 12.9 to 454.4 mg/kg, respectively. In comparison with grade II values of soil environmental quality standard of China (MEPPRC 1995), the average concentrations of Cd and Hg in the study area are 10.0- and 1.7-times greater than the regional safety values, whereas the maximum concentrations of Cd, As, Cu, Hg and Zn are 23.7-, 1.4-, 1.7-, 5.0- and 2.3-times the national standard values, respectively.

| Heavy metal (mg/kg) | Cd  | As  | Cu  | Hg  | Zn  |
|---------------------|-----|-----|-----|-----|-----|
| Min                 | 1.0 | 2.5 | 0.3 | 0.2 | 12.9 |
| Max                 | 7.1 | 40.4| 86.1| 1.5 | 454.4|
| Mean                | 3.1 | 12.3| 31.4| 0.5 | 178.7|
| Standard value (Grade II) | 0.3 | 30  | 50  | 0.3 | 200 |
| Mean fold           | 10.0| —   | —   | 1.7 | —   |
| Maximum fold        | 23.7| 1.4 | 1.7 | 5.0 | 2.3 |

Table 1. Statistics of heavy metal concentrations at 33 locations (at 0–20 cm in depth) in the study area. Note: Heavy metal concentrations more than the secondary criterion are referred to as "soil pollution". Grade II of environmental quality standards values for soils of China (MEPPRC 1995).
Human health risk caused by heavy metals. Figures 2 and 3 show the non-carcinogenic risk (HI) and carcinogenic risk (CR) resulting from human exposure to heavy metal contamination in the study area before remediation. It is clear that the non-carcinogenic risks of the industrial park for Cd, Cu, Hg and Zn in soil are <1.0 (Fig. 2(a,c–e)); for As, is partly >1.0 (Fig. 2(b)). Meanwhile, Fig. 3(a) shows that the carcinogenic risks for As (varying from 1.83E-06 to 5.88E-05) across the entire industrial park are greater than the standard acceptable risk safety level for a single contaminant (1.0E-06); areas with the highest carcinogenic risk cluster in the central industrial park. For Cd contamination, the elevated carcinogenic risk areas cover almost the whole industrial
Figure 2. Spatial distribution of non-carcinogenic risk for each specific heavy metal before remediation: (a) Cd; (b) As; (c) Cu; (d) Hg; (e) Zn (Note: non-carcinogenic diseases might be caused by heavy metal exposure if HI > 1; ArcGIS 10.1 was used to create the map in this figure, http://www.esrichina.com.cn/2015/0107/2830.html).

Figure 3. Spatial distribution of carcinogenic risk before remediation for As (a) and Cd (b) (Unit: 10^-6) (Note: generally cancer might be caused by heavy metal exposure if CR > 1; ArcGIS 10.1 was used to create the map in this figure, http://www.esrichina.com.cn/2015/0107/2830.html).
park, with the highest risk recorded at 5.84E-06. However, there is still a small area with carcinogenic risk under 1.0 in the southeastern corner of the industrial park (Fig. 3(b)).

Figure 4 displays the distribution maps of multiple heavy metals’ CR risk, HI risk and overall risk (i.e., overlaid raster of CR and HI risks), as well as the identified associated contaminated areas. As shown in Fig. 4(a), the contaminated areas where HI risks are higher than the acceptable level (i.e., 1.0) are mainly concentrated in the middle and northern industrial park, accounting for 8.1% of the entire area. Meanwhile, areas with CR risks over the acceptable level (i.e., 1.0E-04) are located in the south-eastern and partly in the western industrial park, with a proportion up to 18.2% (Fig. 4(b)). In addition, the results combined in Fig. 4(c) also indicate that the contaminated areas identified as overall risk areas are concentrated sparsely in the middle residential region and the non-residential region in the northeast, accounting for approximately 26.0% (partly superposed, located in the middle residential region) of the industrial park.

**Contributions of heavy metals to human health risk.** Table 3 presents the contribution of heavy metal concentration in topsoil to health risk before remediation. The influences of single factors on health risk, listed in the order of PD values are: As (0.460) > Cu (0.312) > Zn (0.305) > Cd (0.267) > Hg (0.158). As concentration plays the greatest role in overall risk, followed by Cu, Zn and Cd. Hg contributes slightly to the overall risk. Meanwhile, the joint impacts of two factors reveal the interactive effects between As and Cd (0.682), As and Cu (0.654), As and Hg (0.795), As and Zn (0.620), Cd and Cu (0.600), Cd and Hg (0.547), Cd and Zn (0.611), Cu and Hg (0.679), Cu and Zn (0.593), Hg and Zn (0.697) appear to be stronger than the impacts of the corresponding separate factors. Even those factors with lesser interaction impacts are likely to enhance their separate effects on
human health. However, after interacting Cd with Hg, and Zn, Cu, and Hg with As, as well as Cu with Zn, the relationships between them are bi-linear.

**Reliability analysis of integrated H-G scheme.** Figures 5 and 6 show the concentration surfaces of As and Cd, as well as the associated exposed CRs selected to assess the reliability of the integrated H-G scheme in identifying the heavy metal polluted soils for remediation in study area. Comparing with the results in Fig. 1, it is clear that the relative high concentrations of As and Cd in the area necessary for remediation identified by the integrated H-G scheme are obviously cut down and are close to the corresponding grade II thresholds of soil environmental quality standard of China after remediation. Comparison of Figs 3 and 6 also reveals that the associated CRs in the area are significantly reduced accordingly, and are finally under the acceptable risk thresholds in this area considering the background concentrations of heavy metals.

**Discussion**
This study analyzed the heavy metal contamination of topsoil based on data collected from a renewable resource industrial park in mainland China. In this process, the human health risk assessment model was applied to judge

| Determinants | Cd | Hg | Zn | Cu | As |
|--------------|----|----|----|----|----|
| **PD for single factor** | 0.267 | 0.158 | 0.305 | 0.312 | 0.460 |
| **PD for joint factors** | Cd | Hg | 0.547 | Zn | 0.611 |
| | | | 0.697 | Cu | 0.690 |
| | | | 0.679 | As | 0.682 |
| | | | 0.795 | | 0.620 |
| | | | | | 0.654 |

Table 3. Single factor and joint factors’ detection by Geo-detector.

**Figure 5.** Spatial distribution of the heavy metal concentrations for Cd (a) and As (b) after remediation (Note: ArcGIS 10.1 was used to create the map in this figure, http://www.esrichina.com.cn/2015/0107/2830.html).

**Figure 6.** Spatial distribution of carcinogenic risk for As (a) and Cd (b) after remediation in areas identified by the H-G scheme (Unit: 10⁻⁶) (Note: ArcGIS 10.1 was used to create the map in this figure, http://www.esrichina.com.cn/2015/0107/2830.html).
soil contamination risk and identify contaminated areas that require remediation. The primary pollutant of the total heavy metal contamination was detected using the Geo-detector method. Consequently the reliability of the integrated H-G scheme in identifying the heavy metal polluted soils for remediation was assessed through comparing the concentration surfaces of critical heavy metal pollutants, as well as the associated exposed CRs in the identified areas before and after remediation. The results highlight that the innovative integrated H-G scheme combining human health risk assessment and the Geo-detector methods based on GIS mapping technology is helpful for identifying areas for soil remediation and the primary heavy metal contributors with limited site samples. Meanwhile, compared to the traditional cost intensive and limited representation point sampling strategy, the integrated H-G scheme demonstrates a cost advantage. Using the IDW interpolation method provides a fine resolution soil remediation investigation through a continuously interpolated surface of health risks based on a limited number of site sampling inputs.

The descriptive statistical evidence confirmed high heterogeneity and variability of the heavy metal concentrations in the sampled sites over the industrial park, which might result from current or past anthropogenic sources. As a renewable resource industrial park, the sources of heavy metals in soil are mainly from disassembly of used electronic devices, oil refining from scrap automobile tires, and polluted surface runoff. However, this study highlights several limitations and areas for further study. First, relevant parameters on exposure were based on national standard values. When such information is directly employed at the local situ-
national pilot industrial park of a circular economy in 2005, heavy metal contamination in this area aggregated quickly, especially in the vicinity of working facilities. However, the residential usage in this area is still up to 52.9% because it is an area of craft production, which makes implementing fine-scale health risk assessment and soil remediation especially urgent in this area.

**Sampling and analytic method.** For identifying the heavy metal polluted areas necessary for soil remediation, topsoil (at a depth of 0–20 cm) samples with average distance approximately 250 m at thirty-three sites were taken from the industrial park (Fig. 7), considering the distribution of recycling sources. To assess the reliability of the integrated H-G scheme, samples located at the places with heavy metals’ concentrations exceeding the grade II thresholds of soil environmental quality standard of China were recollected after remediation. Longitudes and latitudes of sampling locations were recorded by GPS receiver. Prior to measurement of heavy metal concentrations, soil samples were digested in a mixture of HF, HNO₃, and HClO₄. Then, we utilized atomic absorption spectrometry (China Standard GB/T 22105.1-2008) to analyze concentrations of Cd, Cu and Zn. Concentrations of As and Hg were measured by an atomic fluorescence spectrometer (China Standard GB/T 17138-1997 and GB/T 17141-1997). Quality assurance and quality control procedures were conducted by using standard reference material (GBW07401-GBW07408). All standard calibrations were prepared in the same acid matrix used for the soil samples. Meanwhile, this study performed the statistical analysis using IBM SPSS Statistics 19.0 for Windows.

**Human health risk assessment and geographical detector methods.** The empirical methodology of this study is composed of three parts: IDW interpolation, Human health risk assessment, and Geo-detector analysis.

**Spatial distribution mapping by IDW interpolation.** To recognize soil contamination from heavy metals across the entire industrial park more explicitly, this study applied the IDW spatial interpolation with ArcGIS (version 10.1) for mapping the spatial patterns of heavy metal concentrations. IDW is commonly used in spatial interpolation and has been introduced into contaminated site assessment. It is a type of deterministic method for multivariate interpolation with a set of known scattered points. The values assigned to unknown points are calculated based on the weighted averages of values available at known points. It applies the inverse distance to each known point when assigning weights, given by
\[
Z = \sum_{i=1}^{n} \frac{1}{(D_i)^p} \sum_{j=1}^{n} \frac{1}{(D_j)^p}
\]

where \(Z\) denotes the value of the interpolation points, \(Z_i (i = 1 \sim n)\) is the value of the sample points; \(n\) denotes the number of calculated sample points; \(D_i\) is the distance from sample point \(i\) to the interpolation point; and \(p\) is a positive power parameter determined by the minimum mean absolute error and significantly influences the outcome of interpolation. Additionally, 'n-1 cross validation' was implemented to ensure the IDW interpolation accuracy in this study.

**Human health risk assessment for heavy metals.** Human health risk assessment is a widely used to assess the potential health risk posed by heavy metals in soils to exposed people over a specified time period. The human health risk assessment model originating from the US EPA (USEPA 2007) has been recommended by the Environmental Protection Agency of China. According to the technical guidelines for risk assessment of contaminated sites in China (HJ/T 25-2014) and generally international environmental safety concerns\(^{13}\), the risks of heavy metals to local residents can be estimated using Eqs (2)–(5).

\[
\text{Risk} = HI + CR = \sum_{i=1}^{n} \sum_{j=1}^{n} CDI_{ij} + \sum_{i=1}^{n} \sum_{j=1}^{n} CDI_{ij} \times SF
\]

\[
CDI_{oral} = \frac{C_{soil} \times IR \times ED \times EF \times CF}{BW \times AT}
\]

\[
CDI_{dermat} = \frac{C_{soil} \times ED \times EF \times CF \times SA \times AF \times ABS}{BW \times AT}
\]

\[
CDI_{particle} = \frac{C_{soil} \times PI \times DA \times ED \times EF \times fs \times CF}{BW \times AT}
\]

where \(HI\) characterizes the total non-carcinogenic risk, and \(CR\) is the overall carcinogenic risk of all toxicants via exposure pathways, including oral ingestion, dermal contact and particle inhalation\(^{17,30}\); \(i\) is one of three exposure routes, ingestion, dermal contact and particle inhalation; \(j\) represents the heavy metal contaminant; and \(CDI\) is the chemical daily intake of a contaminant for an individual (with 70-year as the life cycle), mg/(kg·d); the relevant parameters of the model are listed in Table 4 (HJ/T 25-2014). \(SF\) for As and Cd is 1.50 (mg·kg\(^{-1}\)·d\(^{-1}\))\(^{-1}\) and 0.38 (mg·kg\(^{-1}\)·d\(^{-1}\))\(^{-1}\), respectively.

| Parameters | Meaning and value | Parameters | Meaning and value |
|------------|-------------------|------------|-------------------|
| CDI\(_{oral}\) | CDI via ingestion, mg/(kg·d) | CDI\(_{dermat}\) | CDI via dermal contact, mg/(kg·d) |
| CDI\(_{particle}\) | CDI via particle inhalation, mg/(kg·d) | AF | skin adherence factor, 1, mg·cm\(^{-2}\) |
| IR | ingestion rate of soil, 100, mg/d | BW | body weight, 55.9, kg |
| CF | conversion factor, 10^{-3}, kg·mg | ED | exposure duration, 30, a |
| EF | exposure frequency, 350, d/a | SA | surface area of the skin, 5000, cm\(^2\)/d |
| AT | average time, 365·d·a\(^{-1}\) × 70, d | C\(_{soil}\) | concentration of the exposure contaminant, mg/kg |
| ABS | absorption factor, 0.001 | PI | retention fraction of inhaled particulates in body |
| DA | daily air inhalation rate, m\(^3\)/d | f\(_s\) | fraction of soil-borne particulates |
| SF | slope factor, kg·d/mg | RfD | reference dose, mg/(kg·d) |

**Table 4.** Parameters employed for assessing human exposure risks.
Geo-detector analysis for predominant contaminants. As mentioned in the 'Introduction' section, based on spatial consistency of variables, Geo-detector was introduced to detect the main contaminant. In this study, the 'Factor detector' and 'Interaction Detector' were used. The calculation formula of its grounded PD is shown in Eq. (6).

$$PD = 1 - \frac{1}{N\sigma^2} \sum_{i=1}^{L} N_i \sigma_i^2$$

(6)

The whole area N designed to calculate PD is stratified into L strata, denoted by i = 1, ..., L according to the concentration classification of heavy metals, defined as an attribute (the argument), whose statistical properties (e.g., mean and standard deviation) change over space. In Eq. (5), N and $\sigma^2$ denote the area and variance of the dependent variable, respectively, for each i stratum; N represents the whole area. PD $\in [0, 1], PD = 1$ means heavy metal concentration completely controls the overall risk, whereas PD = 0 means the concentration is completely unrelated to overall risk.

In this study, we first classified the overall health risk and heavy metal concentrations using a default interval classification and then loaded the distribution layers of all influential contaminants and the overall health risk into ArcGIS 10.1. After intersecting all layers, factor attributes of these layers were extracted and input into the Geo-detector model. The threshold for statistical significance was determined at $p = 0.05$. In this process, the overall health risk obtained by overlaying the carcinogenic and non-carcinogenic risk layers in ArcGIS 10.1 was employed as a dependent variable; each metal concentration in soil was taken as an independent variable to analyzes the contribution of pollutants, Cd, As, Cu, Hg and Zn, to the total health risk level.

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**Author Contributions**

B.Z. conceived the experiment(s) and wrote the paper, X.J., and J.Z. conducted the experiment(s), B.Z., X.J., X.D., G.S., and X.Z. analyzed the results, X.J., J.Z., J.T. generated the maps, and all authors reviewed the manuscript.

**Additional Information**

**Competing Interests:** The authors declare that they have no competing interests.

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