Distribution Characteristics, Pollution Assessment, and Source Identification of Heavy Metals in Soils Around a Landfill-Farmland Multisource Hybrid District

Honghua Liu¹ · Yuan Wang² · Jie Dong¹ · Lixue Cao¹ · Lili Yu¹ · Jia Xin¹,²

Received: 8 January 2021 / Accepted: 18 May 2021 / Published online: 31 May 2021
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Abstract
Heavy-metal pollution is a negative impact of municipal solid-waste landfills. The multiple pollution transport pathways (including leachate, runoff, and waste gas) and complex and co-existing potential pollution sources (such as agricultural activities) around landfills require a combination of different pollution assessment methods and source identification tools to address pollution distribution and potential risks. In this study, the distributions of eight heavy metals (chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), arsenic (As), cadmium (Cd), and mercury (Hg)) around a landfill were analyzed using 60 topsoil samples. Ecological risk assessments indicated that there are currently no ecological risks. Based on health risk assessments, however, high concentrations of Cr and As in the soil pose a noncarcinogenic and carcinogenic risk to humans in the study area, respectively. In addition, the geoaccumulation indices for Cr, Cu, Ni, Zn, As, and Hg confirmed anthropogenic sources of accumulation of these metals in soils. Additionally, the potential ecological risk index indicated that Hg posed a considerable risk to the ecology of the area around the landfill. Sources of heavy metals in the study area were attributed to natural sources (22.10%), agricultural activities (27.65%), landfill (31.35%), and transportation (18.89%). The continuous accumulation of heavy metals and health risk for humans suggests the need to continuously monitor of heavy metal content and migration around the landfill. This study provides a reference for local authorities in the study area.
reported accumulation of heavy metals in soil, water, and plants near landfills, which verified the potential environmental impacts of landfills. Researchers have suggested environmental management measures to reduce environmental risks in polluting and nonpolluting landfills (Zhou et al. 2015; Ogubanjo et al. 2016; Wang et al. 2020). Closed landfills tend to contain higher concentrations of heavy metals than open landfills (Adelopo et al. 2018). Moreover, heavy metal pollution often is greater in landfills that have been in operation for longer periods (Chang et al. 2007). Due to its affinity for heavy metals, higher organic matter content could contribute to decreased heavy metal leaching in landfills (Cao et al. 2013). These information demonstrated that the distribution characteristics and transport behaviors of heavy metals in soil around landfills have already been investigated in many previous studies.

Landfill sites are not usually isolated and often are adjacent to farmland and factories. Consequently, heavy metal pollution around landfill sites involves a combination of multiple sources. The accumulation of heavy metals in soils around the largest landfill in China is associated with landfill leachate and agricultural activities (Liu et al. 2013). However, most existing studies tend to assess heavy metal pollution in or around landfill sites, without considering the potential impacts of other pollution sources (Bakis and Tuncan 2011; Liu et al. 2013). Our study evaluated heavy-metal pollution under the effect of complex pollution sources, including the landfill site and farmland, which can provide a reference for local management and subsequent site evaluation. In this study, we applied various methods to search for the origin and to assess the pollution potential of eight important heavy metals in soils around a landfill-farmland multisource hybrid district. The positive matrix factorization (PMF) model effectively can realize source identification and apportionment and has been widely applied to farmland (Guan et al. 2018; Gan et al. 2019; Xiao et al. 2019) and industrial sites, including factories (Xue et al. 2014), industrial estates (Zhou et al. 2016; Zhang et al. 2018), coal mines (Bilal et al. 2019; Cheng et al. 2020), and coal-mining cities (Liang et al. 2017; Sun et al. 2019). In this study, we employed the PMF model for source apportionment in sites subjected to the combined effects of landfill- and farmland-generated contamination. In addition, the Spearman correlation coefficient and spatial distribution were used to aid in source identification analysis.

The present study was conducted at a landfill serving the city of Qingdao, China that receives 4300 tonnes of municipal solid waste daily, and which is suspected to pose a potential threat to the surrounding environment. In addition, villages and farmlands surround the landfill. Heavy metal distributions in the soil may thus be affected by multiple sources. Therefore, evaluating the soil quantity around the landfill and identifying the dominant pollution sources and pollution transport pathways are necessary. The goal of this study was to investigate the contamination and risks posed by chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), arsenic (As), cadmium (Cd), and mercury (Hg) in soils near the landfill to analyze the distribution characteristics of these elements and to identify sources and transport pathways of heavy metals in the study area.

Materials and Methods

Study Area

All samples were collected around a landfill, located in Qingdao, Shandong Province, China. The landfill collects 4,300 t/d of municipal solid waste from the six principal urban districts of Qingdao and was established in 2011. As the amount of waste increasing, the landfill was expanded in 2017 by introducing more environmental protection facilities and waste disposal facilities, such as waste incinerators. The land-use types of the study area included farmland, woodland, residential area with livestock farms, and landfill, accounting for approximately 50%, 10%, 10%, and 30%, respectively. No industries apart from the landfill coexisted in the study area at the time of investigation. The weather in Qingdao is characterized by a mean temperature of 13.3 °C, annual precipitation of 722.7 mm, and the dominant wind direction is south–southeast, with highest winds in the spring and summer.

Sample Collection and Laboratory Analysis

In June 2019, we collected 60 soil samples within a radius of 1 km from the landfill. The southeastern part of the landfill is separated by a river and not covered in our study. Figure 1 shows a map of the study area and the sampling site distribution. To better represent variations in the spatial distributions of the elements, the sampling sites were divided into three groups according to their locations relative to the landfill site (Fig. 1): Group 1 (eastern area), Group 2 (northern area), and Group 3 (western area). Of the total samples, 11, 15, and 34 belonged to Groups 1, 2, and 3, respectively.

A grid sampling method was used for the layout of the sampling sites. The sample (approximately 1.5 kg) for an individual site was composed of five subsamples taken from the topsoil (0–20 cm) within a 20–50 m radius. Roots, stalks, stones, insect bodies, and other sundry materials were discarded. After air-drying and sifting, the samples were sent for laboratory analysis.

Soil pH was determined using a pH meter (MAPRC 2006a), and the organic matter (OM) content was measured using oil bath-K2Cr2O7 titration (MAPRC 2006b). X-ray fluorescence spectrometry (XFP; Axiosmax, Panak,
Fig. 1  Location of the study area and distribution of sampling sites (SS)
Netherlands) was used to measure the concentrations of Ni, Cu, Pb, Zn, and Cr (Lu et al. 2010; Kodom et al. 2012; Tang et al. 2015). Atomic fluorescence spectrometry (AFS; AFS-9750, Beijing Haiguang Instrument, China) was used to determine the Hg and As concentrations (Yang et al. 2011; Tan et al. 2014; Wang et al. 2015). Cd concentrations were measured by inductively coupled plasma mass spectrometry (ICP-MS, NexION 2000, PerkinElmer, USA) (Hu et al. 2013; Tang et al. 2015; Huang et al. 2020). For quality control (QC) and quality assurance (QA), all measurements were conducted at the Shandong Institute of Geophysical and Geochemical Exploration and Prospecting, which passed the China Metrology Accreditation. To ensure quality control, reagent blanks, duplicate samples, and certified reference standard soil samples (GBW07309, GBW07425, and GBW07451) were included in the analytic process. The relative deviation (RD) was maintained within 25%. The detection limits of Cr, Pb, Cu, Zn, Ni, Cd, Hg, and As were 3.0, 2.0, 1.2, 2.0, 1.5, 0.07, 0.002, and 0.01 mg/kg, respectively.

Assessment of Heavy Metal Contamination

In this study, a combination of multiple assessment methods, including pollution, geo-accumulation, and potential ecological risk indexes, was utilized to evaluate heavy-metal contamination.

The single pollution index (PI) refers to the pollution level of each heavy metal in the soil, and the Nemerow integrated pollution index (NIPI) indicates the total heavy metal pollution level in the study area. The PI values for individual elements (PIi) and NIPI were calculated according to Eqs. (1) and (2), respectively:

\[ PI_i = \frac{C_i}{C_{b,i}} \]  
\[ \text{NIPI} = \sqrt{\left(\frac{PI_i}{\text{PI}_{\text{max}}}\right)^2 + \left(\frac{PI_{\text{max}}}{2}\right)^2} \]  

where \( C_i \) indicates the content of each individual element (mg kg\(^{-1}\)), \( C_{b,i} \) indicates the corresponding value specified in the Soil environment quality Risk control standard for soil contamination of agricultural land (mg kg\(^{-1}\)) (MEEPRC 2018), \( \text{PI}_{\text{max}} \) indicates the mean PI value (unitless), and \( \text{PI}_{\text{max}} \) indicates the maximum PI value (unitless). The classification criteria for PI and NIPI are summarized in Table S1 in Supplementary Information (Nemerow 1985; Fei et al. 2019).

The geo-accumulation index (\( I_{\text{geo}} \)), originally used for sediments, is a widely used assessment method for revealing the enrichment of elements in soil (Loska et al. 2004) that can be calculated using Eq. (3):

\[ I_{\text{geo}} = \log_2 \left( \frac{C_i}{1.5C_{b,i}} \right) \]  

where \( C_i \) represents the content of the individual metal element (mg kg\(^{-1}\)), and \( C_{b,i} \) represents the background value (mg kg\(^{-1}\)), where the values are Cr = 47.78, Cu = 18.5, Ni = 18.63, Pb = 29.630, Zn = 56.68, As = 5.76, Cd = 0.139, and Hg = 0.035 (Zhang 2011). Table S2 in Supplementary Information divides \( I_{\text{geo}} \) into seven levels (Muller 1969).

Potential ecological risk is usually applied to estimate the effects of heavy metals on an ecological system. The ecological risk index (ER) of each element was calculated using Eq. (4). The overall potential ecological risk index (PER) is given as the sum of all ERs.

\[ ER_i = T_i \times \frac{C_i}{C_{b,i}} \]  
\[ \text{PER} = \sum_{i=1}^{n} ER_i \]  

Health-Risk Assessment

Health risks for both adults and children were estimated in the study area. The exposure pathways of metal elements primarily comprised ingestion, inhalation, and dermal contact, which are herein referred to as \( D_{\text{ing}}, D_{\text{inh}}, \) and \( D_{\text{dermal}} \) (mg kg\(^{-1}\) d\(^{-1}\)), respectively. The principal ingestion pathway was the consumption of vegetables in the study area. The concentrations of heavy metals in vegetables and the mean daily doses received through individual exposure pathways were calculated using Eqs. (6), (7), (8), and (9) (US EPA 1989; Wang et al. 2005; US EPA 2015):

\[ C_{\text{vegetable}} = C_{\text{soil}} \times TF_i \]  
\[ D_{\text{ing}} = \frac{C_{\text{vegetable}} \times IngR_{\text{vegetable}} \times EF \times ED}{BW \times AT} \]  
\[ D_{\text{dermal}} = \frac{C_{\text{soil}} \times SL \times CF \times SA \times ABS \times ED \times ED}{BW \times AT} \]
\[ D_{\text{inh}} = C_{\text{soil}} \times \frac{\text{InhR} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BW} \times \text{AT}} \]  

where \(C_{\text{vegetable}}\) represents the content of the individual metal element in vegetables (mg kg\(^{-1}\)), \(C_{\text{soil}}\) represents the content of the individual metal element in soil (mg kg\(^{-1}\)), \(\text{TF}_i\) represents the ratio of element contents in vegetables to soil (unitless), \(\text{IngR}_{\text{vegetable}}\) represents the individual ingestion rate of vegetable (mg d\(^{-1}\)), \(\text{CF}\) represents the conversion factor applied in mathematical calculations (kg mg\(^{-1}\)), \(\text{EF}\) represents individual exposure frequency to the pollutant (year), \(\text{BW}\) represents mean body weight (kg), \(\text{AT}\) represents the mean individual exposure time (days), \(\text{SL}\) represents the skin adherence factor (mg cm\(^2\) h\(^{-1}\)), \(\text{SA}\) represents the mean skin area (cm\(^2\)), \(\text{ABS}\) represents the dermal absorption factor (unitless), \(\text{InhR}\) represents the inhalation rate of soil (m\(^3\) d\(^{-1}\)), and \(\text{PEF}\) represents individual exposure frequency to the pollutant (year), \(\text{BW}\) represents mean body weight (kg), \(\text{AT}\) represents the mean individual exposure time (days), \(\text{SL}\) represents the skin adherence factor (mg cm\(^2\) h\(^{-1}\)), \(\text{SA}\) represents the mean skin area (cm\(^2\)), \(\text{ABS}\) represents the dermal absorption factor (unitless), \(\text{InhR}\) represents the inhalation rate of soil (m\(^3\) d\(^{-1}\)), and \(\text{PEF}\) represents the particle emission factor (mg cm\(^2\) h\(^{-1}\)). The values of the parameters are summarized in Table S4 (US EPA 2001; Wang et al. 2005; Zheng et al. 2010) and S5 (Bian et al. 2014; Hu et al. 2014) in Supplementary Information.

The noncarcinogenic risk was estimated by Eq. (10), and the carcinogenic risk was calculated with Eq. (11) (US EPA 1989):

\[ \text{HI} = \sum HQ_i = \sum \frac{D_i}{\text{RfD}_i} \]  

\[ \text{Risk}(\text{RI}) = \sum D_i \times \text{SF}_i \]  

where \(\text{RfD}_i\) indicates the reference dose for a single heavy metal (mg kg\(^{-1}\) d\(^{-1}\)), and \(\text{SF}_i\) indicates the carcinogenic slope factor for a single heavy metal (mg kg\(^{-1}\) d\(^{-1}\)). Table S5 in Supplementary Information summarizes the values of the parameters (MEEPRC 2014; Xiao et al. 2015; Pan et al. 2016; Eziz et al. 2018).

**Source Identification**

The Positive Matrix Factorization (PMF) model is recommended by the United States Environmental Protection Agency (US EPA) for source identification (Norris et al. 2014). Equation (12) shows that the concentration data matrix \(X\) can be represented by three matrices: one each for factor contribution \((G)\), source profile \((F)\), and residual error \((E)\) (Jiang et al. 2017):

\[ X_{ij} = \sum_{k=1}^{p} G_{ik} F_{jk} + E_{ij} \]  

where the subscripts \(i, j\), and \(p\) represent the number of samples, elements, and factors, respectively. After minimizing the objective function \(Q\), \(E_{ij}\) is obtained as follows:

\[ Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left( \frac{E_{ij}}{\mu_{ij}} \right)^2 \]  

where \(\mu_{ij}\) represents uncertainty in the concentration values. In this study, the uncertainties were calculated by using Eq. (14):

\[ \mu_{ij} = 0.1x_{ij} + \frac{\text{MDL}}{3} \]  

where \(x_{ij}\) represents the content of element \(j\) in sample \(i\), and \(\text{MDL}\) represents the method detection limit.

With inputs of data on the concentrations of metals in the samples and the level of uncertainty of the sample data, the PMF model can provide outputs that can be used for source identification of the metal pollutants.

**Data Analysis**

Spearman correlation coefficients were applied in the analysis of data with a nonnormal distribution. Microsoft Excel 2016 and SPSS version 25 were used for statistical analyses. The spatial distributions of metal elements were obtained using ArcGIS software version 10.2, using the inverse distance weighted (IDW) method. PMF version 5.0 was used to identify potential pollution sources.

**Results and Discussion**

**Soil Properties**

Table S6 in Supplementary Information shows the statistical data for soil properties, including the pH value and OM of soil around the landfill. The acidic (pH = 5.77) soil could increase the release of heavy metals (Chai et al. 2007). The soil samples from the study area contained higher contents of OM (2.15%) than the background concentration (CNEMC 1990).

**Heavy metal concentrations**

The detected concentrations of Cd, Cu, Pb, Ni, As, and Hg in the three certified reference materials were 0.27, 35, 22, 33, 7.1, and 0.019 mg kg\(^{-1}\), respectively. The standard values and uncertainty for Cd, Cu, Pb, Ni, As, and Hg were 0.26 ± 0.04, 32 ± 2, 23 ± 3, 32 ± 2, 7.4 ± 0.5, and 0.020 ± 0.002 mg kg\(^{-1}\), respectively. Table 1 illustrates the statistical data of metal element concentrations in soils around the studied landfill site compared with concentrations...
Table 1  Summary statistics of heavy metal concentrations (mg kg\(^{-1}\)) in the soils around the landfill and comparison landfills

|        | Cr    | Cu    | Ni    | Pb    | Zn    | As    | Cd    | Hg    |
|--------|-------|-------|-------|-------|-------|-------|-------|-------|
| Min    | 53.58 | 16.69 | 12.82 | 20.41 | 47.87 | 7.46  | 0.07  | 0.03  |
| Mean ± SE\(^{a}\) | 67.77 ± 1.35 | 23.31 ± 0.54 | 27.62 ± 0.83 | 24.71 ± 0.29 | 63.07 ± 1.41 | 10.77 ± 0.24 | 0.10 ± 0.00 | 0.05 ± 0.00 |
| Max    | 123.76 | 30.71 | 53.79 | 29.71 | 109.98 | 15.99 | 0.14  | 0.12  |
| SD\(^{b}\) | 10.48 | 4.15  | 6.46  | 2.22  | 10.92 | 1.88  | 0.02  | 0.01  |
| CV\(^{c}\) | 0.15  | 0.18  | 0.23  | 0.09  | 0.17  | 0.18  | 0.15  | 0.29  |
| Background value | 47.48 | 18.50 | 18.63 | 29.63 | 56.69 | 5.76  | 0.14  | 0.035 |
| Chinese soil guideline\(^{d}\) (pH ≤ 5.5) | 150   | 50    | 60    | 70    | 200   | 40    | 0.3   | 1.3   |
| Chinese soil guideline\(^{d}\) (5.5 ≤ pH ≤ 6.5) | 150   | 50    | 70    | 90    | 200   | 40    | 0.3   | 1.8   |
| Chinese soil guideline\(^{d}\) (6.5 ≤ pH ≤ 7.5) | 200   | 100   | 100   | 120   | 250   | 30    | 0.3   | 2.4   |

Landfill Country Operation time (years) Disposal capacity (t/d)

| Reference [1] | Laos | 12  | 300 | 14.68 | 13.65 | 7.63 | 18.75 | 4.59 | -   | 1.04 | -   |
| Reference [2] | Czech | 21  | 37.5| 77.16 | 48.41 | 41.47 | 4.87  | 35.25 | -   | -   | 0.04 |
| Reference [3] | China | 23  | 5,000 | 86.00 | 34.00 | -    | 11.60 | 99.50 | -   | 0.20 | -   |
| Reference [4] | Italy | 22  | -   | 89.90 | 25.50 | 54.80 | 22.70 | 87.20 | 5.60 | 0.31 | 0.03 |

\(^{a}\)Standard error  
\(^{b}\)Standard deviation  
\(^{c}\)Coefficient of variation  
\(^{d}\)Soil environment quality risk control standard for soil contamination of agricultural land

[1] (Vongdala et al. 2019), [2] (Adamcova et al. 2017), [3] (Liu et al. 2013), [4] (Nannoni et al. 2017)
at other landfill sites. Generally, the mean concentrations of Cr, Zn, Ni, Pb, Cu, As, Cd, and Hg were 67.77, 63.07, 27.62, 24.71, 23.31, 10.77, 0.10, and 0.05 mg kg\(^{-1}\), respectively. Table 1 shows that the heavy metals in this study did not cause soil contamination and had concentrations lower than the risk screening values of agricultural land in China (MEEPRC 2018). However, the mean contents of most metals (Cr, Cu, Ni, Zn, As, and Hg) were significantly higher than those near a landfill in Vientiane, Laos of 268 t/d (Vongdala et al. 2019), which might be a result of the shorter time of operation of the landfill (contaminating 81.67% of the samples) and seriously influenced by unnatural factors.

Among the eight elements studied, Hg had the highest coefficient of variation (CV 0.29), which was associated with anthropogenic activities (Jiang et al. 2017). Table 1 reveals that the average contents of Cr, Cu, Ni, Pb, and Zn (67.77, 23.32, 27.62, 24.71, and 63.07 mg kg\(^{-1}\), respectively) in soils around the studied landfill (4,300 t/d) were significantly higher than those near a landfill in Vientiane, Laos of 268 t/d (Vongdala et al. 2019), which might be associated with the greater daily waste disposal capacity of the landfill studied herein. In addition, soil near the studied landfill (8 years in operation) contained lower concentrations of Cr, Cu, and Ni than soils around a municipal solid waste landfill in Pilsen, Czech Republic (21 years in operation) with concentrations of Cr, Cu, and Ni of 77.16, 48.41, and 41.47 mg kg\(^{-1}\), respectively (Adamcova et al. 2016), the Laogang landfill in Shanghai, China (23 years in operation) with concentrations of Cr and Ni of 86 and 34 mg kg\(^{-1}\) for Cr, respectively (Liu et al. 2013), and the Ginestreto municipal solid waste landfill in Italy (22 years in operation) with concentrations of Cr, Cu, and Ni of 89.0, 25.5, and 54.8 mg kg\(^{-1}\), respectively (Nannoni et al. 2017). The lower concentrations of metals in the present study might be a result of the shorter time of operation of the landfill. A previous study (Chang et al. 2007) reported that heavy-metal pollution tends to be more serious in landfills that have been operating for longer periods of time. The higher concentrations of Pb and As in the landfill under study than in the three landfill samples with longer operating times were perhaps associated with the specific waste composition (Ishchenko 2019).

### Heavy-Metal Contamination Assessment

#### Pollution Index

Table S7 in Supplemental Information illustrates the assessment results, including pollution classes for the topsoil samples. The PI values for eight of the studied metal elements were < 1, demonstrating that heavy metals for all the samples were in the safe class and posed little threat to the quality and safety of agricultural production. In addition, the NIPI value (0.40) also suggests that there was no environmental pollution caused by heavy metals.

#### Geo-accumulation index

\( I_{geo} \) was calculated to estimate the levels of heavy metal accumulation in soils under the influence of human activities. Table S8 in Supplementary Information summarizes the pollution level classification according to the \( I_{geo} \) evaluation results. The percentages of samples ranging from the uncontaminated to moderately contaminated class were 31.67%, 20.00%, 38.33%, 1.67%, 81.67%, and 33.33% for Cr, Cu, Ni, Zn, As, and Hg, respectively. The varying accumulation of heavy metals in the soil could be attributed to different factors. As was heavily enriched in soils around the landfill (contaminating 81.67% of the samples) and seriously influenced by unnatural factors.

#### Potential Ecological Index

In this study, ER and PER were calculated to estimate ecological risk and are illustrated in Table S9 in Supplementary Information. The average ER was the highest for Hg, followed by Cd, As, Ni, Cu, Pb, Cr, and Zn. The heavy metals (excluding Hg) posed low risks to the surrounding environment, whereas 86.67% of the samples posed considerable risk in terms of Hg content. Thus, Hg should be considered a priority pollutant. According to the categorization standard (Table S3 in Supplementary Information), the mean PER value (117.21) indicated that the ecological environment near the landfill site was at low risk for heavy-metal pollution.

#### Health-Risk Assessment

Table 2 summarizes the risks posed to human health by the eight heavy metals in the soil via various exposure pathways, which generally followed a similar order in terms of risk level for most heavy metals for both adults and children. However, the risk levels of Cr for adults and children were significantly different. In addition to Cr, the HI values decreased in the order of As > Cd > Ni > Pb > Cu > Zn > Hg. The HI values for Cu, Ni, Pb, Zn, As, Cd, and Hg for adults (6.33 × 10\(^{-2}\), 1.57 × 10\(^{-3}\), 1.05 × 10\(^{-3}\), 6.22 × 10\(^{-2}\), 3.59 × 10\(^{-1}\), 2.86 × 10\(^{-1}\), and 4.89 × 10\(^{-3}\), respectively) were lower than the corresponding values for children (1.48 × 10\(^{-1}\), 3.68 × 10\(^{-1}\), 2.45 × 10\(^{-1}\), 1.46 × 10\(^{-1}\), 1.08 × 10\(^{-3}\), 8.33 × 10\(^{-1}\), 6.70 × 10\(^{-1}\), and 1.13 × 10\(^{-2}\), respectively). Similar findings were reported in previous studies (Chen et al. 2019; Baltas et al. 2020; Zhao et al. 2020). Ingestion, inhalation, and dermal contact are the exposure pathways to the heavy metals.
contaminated in the soils. Because the principal land-use type of soils around the landfill site was cropland planted with vegetables, vegetable intake was considered the primary ingestion method. Cr had the highest and second lowest HI values for adults and children, respectively. In addition, the HI value for Cr was higher for adults (5.57) than for children (1.31 × 10⁻⁷); the difference could be due to the relatively high concentration of Cr in soil or to the generally larger consumption of vegetables by adults (0.345 kg d⁻¹) than by children (0.2315 kg d⁻¹). In addition, adults tend to consume more heavy metals by dermal contact because of greater skin area. Metals can be absorbed by the skin into the blood and accumulate in the body (Borowska and Brzoska 2015).

Previous studies have found human health problems posed by heavy metals due to dermal contact (Singh et al. 2018) and inhalation (Xu et al. 2020). Our study results were opposite to those of these studies, perhaps because heavy metals concentrations in this study were not high enough to cause human health problems by dermal contact and inhalation. The hazard quotient (HQ) for each pathway decreased in the order of HQing > HQdermal > HQinh. Thus, ingestion was the dominant exposure pathway for heavy metals in this study. HI values of Cr higher than 1 denotes exposure to heavy metals that could cause noncancer-related health problems for adults in the study area.

The carcinogenic risks were estimated for certain heavy metals (Cr, Ni, As, and Cd) using the SF value. The carcinogenic risks posed to both populations (adults and children) followed the same order of As > Cr > Ni > Cd. The cancer risks posed by As, Cr, Ni, and Cd to adults were 6.87 × 10⁻⁵, 5.66 × 10⁻⁷, 2.09 × 10⁻⁹, and 5.66 × 10⁻¹¹, respectively, which were higher than those posed to children (1.12 × 10⁻⁴, 1.57 × 10⁻⁷, 1.28 × 10⁻⁹, and 3.47 × 10⁻¹¹, respectively). In addition to As, these results corroborate those reported in a previous study that also stated that adults are subject to greater cancer risks than children (Yang et al. 2018). The heavier average weight of adults (70 kg) than of children (20 kg) contributes to higher cancer risk posed by As for children through ingestion. The carcinogenic risks posed by As to humans were higher than 1.00 × 10⁻⁶, indicating that significant health problems are expected (Fryer et al. 2006).

### Spatial Distribution Characteristics and Source Identification of Heavy Metals

Spatial distribution, Spearman correlation coefficients, and the PMF model were applied for an accurate analysis of potential sources of heavy metals in the soils of the study area. Table 3 summarizes the comparison between the three groups of sampling sites based on multiple indexes, including Csoil, PI, NIPI, Igeo, ER, and PER. The index values for Cu, Pb, As, and Hg were highest in Group 2, followed by Group 3 and Group 1.

According to Table S10 in Supplementary Information, the health risks posed by Cu, Pb, As, and Hg followed the same order. Moreover, the Igeo of As and Hg in Group 2 were almost 4 times those of Group 1. Overall, heavy metal contamination was relatively higher in Groups 2 and 3 than in Group 1. Figures 2 and 3 show the distribution maps of the metal elements estimated using IDW methods with GIS technology. Spatially distributed hotspots usually represent high levels of pollution (Doyi et al. 2018). Figures 2 and 3 reveal two hotspots located in the northwestern (Group 2) and southwestern (Group 3) areas around the landfill, which is consistent with the above-presented results. Because a hotspot in the northwestern area was located in a zone impacted by the prevailing southeast wind, the distribution of heavy metals might be associated with atmospheric deposition, as has been reported in previous studies (Li et al. 2017; Jin et al. 2019; Mentese et al. 2021). Due to increased exposure to atmospheric deposition, heavy metal concentrations tend to increase with altitude (Cao et al. 2013). However, the hotspot in the southwestern area is at a low elevation. Surface runoff was suspected to be another dominant transport pathway for heavy metals. Additionally, hotspots roughly corresponded to the positions of the two villages and various agricultural activities. Thus, the overall heavy metal

| Element | Adults | Children |
|---------|--------|----------|
|         | HQing  | HQdermal | HQinh | HI = ΣHQi | Cancer risk | HQing  | HQdermal | HQinh | HI = ΣHQi | Cancer risk |
| Cr      | 5.57E+00 | 1.29E-03 | 2.37E-04 | 5.57E+00 | 2.56E-07 | 1.31E+01 | 6.33E-04 | 2.08E-04 | 1.31E+01 | 1.57E-07 |
| Cu      | 6.32E-02 | 1.11E-04 | 1.22E-07 | 6.33E-02 | 1.48E-01 | 2.56E-07 | 5.44E-05 | 1.29E-06 | 2.45E-01 | 2.56E-07 |
| Ni      | 1.57E-01 | 2.92E-04 | 2.82E-07 | 1.57E-01 | 1.29E-06 | 1.31E-01 | 1.46E-01 | 2.94E-05 | 1.46E-01 | 2.94E-05 |
| Pb      | 1.04E-01 | 2.68E-04 | 1.48E-06 | 1.05E-01 | 1.29E-06 | 2.45E-01 | 1.31E-01 | 1.46E-01 | 2.45E-01 | 1.46E-01 |
| Zn      | 6.22E-02 | 5.99E-05 | 4.42E-08 | 6.22E-02 | 1.46E-01 | 2.94E-05 | 1.46E-01 | 2.94E-05 | 1.46E-01 | 2.94E-05 |
| As      | 3.54E-01 | 4.99E-03 | 7.30E-06 | 3.59E-01 | 1.29E-06 | 8.33E-01 | 1.12E-04 | 8.33E-01 | 1.12E-04 | 8.33E-01 |
| Cd      | 2.85E-01 | 5.69E-04 | 2.10E-08 | 2.86E-01 | 2.79E-04 | 5.69E-01 | 1.83E-08 | 5.69E-01 | 1.83E-08 | 5.69E-01 |
| Hg      | 4.75E-03 | 1.31E-04 | 1.18E-07 | 4.89E-03 | 1.43E-07 | 1.12E-02 | 4.34E-05 | 1.03E-07 | 1.12E-02 | 4.34E-05 |
contamination may be due to a combination of landfill and agriculture.

Spearman correlation coefficients were used to investigate the correlations among the eight heavy metals. As listed in Table S11 in Supplemental Information, Cr was positively correlated with Ni ($r = 0.955$). In addition, Cu exhibited a strong positive correlation with Zn ($r = 0.919$). Thus, the two elements in these pairs may have originated from similar sources.

The PMF model provided more supportive evidence for the identification and apportionment of potential heavy metal sources. The concentration data for 60 samples and

### Table 3

Comparison of three groups of sampling sites based on the mean values of heavy metal concentration ($C_{soil}$, mg/kg), pollution indices (PI and NIPI), geo-accumulation index ($I_{geo}$), and potential ecological risk indices (ER and PER)

|                  | Cr  | Cu  | Ni  | Pb  | Zn  | As  | Cd  | Hg  | Integrated index |
|------------------|-----|-----|-----|-----|-----|-----|-----|-----|------------------|
| **C$_{soil}$**   |     |     |     |     |     |     |     |     |                  |
| Group 1          | 66.84 | 23.12 | 28.49 | 24.97 | 62.66 | 9.66 | 0.10 | 0.04 |
| Group 2          | 67.26 | 23.46 | 26.81 | 25.00 | 61.87 | 12.25 | 0.09 | 0.06 |
| Group 3          | 68.30 | 23.30 | 27.70 | 24.50 | 63.73 | 10.47 | 0.10 | 0.05 |
| **PI**           |     |     |     |     |     |     |     |     |                  |
| Group 1          | 0.45 | 0.46 | 0.44 | 0.31 | 0.31 | 0.24 | 0.34 | 0.03 | 0.41 |
| Group 2          | 0.45 | 0.47 | 0.43 | 0.34 | 0.31 | 0.31 | 0.30 | 0.04 | 0.41 |
| Group 3          | 0.43 | 0.40 | 0.38 | 0.28 | 0.30 | 0.28 | 0.34 | 0.03 | 0.39 |
| **$I_{geo}$**    |     |     |     |     |     |     |     |     |                  |
| Group 1          | -0.10 | -0.29 | 0.00 | -0.84 | -0.46 | 0.14 | -1.01 | -0.45 |            |
| Group 2          | -0.09 | -0.25 | -0.07 | -0.83 | -0.47 | 0.48 | -1.24 | 0.00 |            |
| Group 3          | -0.08 | -0.28 | -0.06 | -0.87 | -0.44 | 0.26 | -1.03 | -0.16 |            |
| **ER**           |     |     |     |     |     |     |     |     |                  |
| Group 1          | 2.82 | 6.25 | 7.65 | 4.21 | 1.11 | 16.78 | 22.33 | 44.91 | 106.05 |
| Group 2          | 2.83 | 6.34 | 7.20 | 4.22 | 1.09 | 21.27 | 19.41 | 63.17 | 125.53 |
| Group 3          | 2.88 | 6.30 | 7.43 | 4.14 | 1.12 | 18.18 | 22.22 | 54.88 | 117.15 |
| **NIPI**         |     |     |     |     |     |     |     |     |                  |
| Group 1          | 0.45 | 0.46 | 0.44 | 0.31 | 0.31 | 0.24 | 0.34 | 0.03 | 0.41 |
| Group 2          | 0.45 | 0.47 | 0.43 | 0.34 | 0.31 | 0.31 | 0.30 | 0.04 | 0.41 |
| Group 3          | 0.43 | 0.40 | 0.38 | 0.28 | 0.30 | 0.28 | 0.34 | 0.03 | 0.39 |
| **PER**          |     |     |     |     |     |     |     |     |                  |
| Group 1          | 2.82 | 6.25 | 7.65 | 4.21 | 1.11 | 16.78 | 22.33 | 44.91 | 106.05 |
| Group 2          | 2.83 | 6.34 | 7.20 | 4.22 | 1.09 | 21.27 | 19.41 | 63.17 | 125.53 |
| Group 3          | 2.88 | 6.30 | 7.43 | 4.14 | 1.12 | 18.18 | 22.22 | 54.88 | 117.15 |

**Fig. 2** Spatial distributions of the concentrations of: **a** Cr; **b** Ni; **c** Cu; **d** Zn
the uncertainties were input into the model. The minimum and most stable value of $Q$ indicate four factors (Fig. 4). Factor 1 was controlled by Cr and Ni with high factor loadings (33.1% and 41.5%, respectively) and corresponds with the distribution characteristics and Spearman correlations, further suggesting the same source for Cr and Ni. The distribution of Cr and Ni was previously reported to be dominated by natural sources, such as parent rocks (Lu et al. 2012; Ke et al. 2017; Zhang et al. 2018). Thus, factor 1 was proposed to be related to a natural source. Factor 2 was dominated by Cu and Zn (loadings of 35.6% and 39.1%, respectively). Our survey revealed nearby private and centralized livestock farms that discharged wastewater and solid wastes generated by farming activities directly into the soil, causing accumulation of Cu and Zn (Cang et al. 2004). Moreover, pesticides and fertilizers are commonly used in farmland in the study area, and excessive application of phosphate fertilizers leads to the enrichment of Zn in soil (Ke et al. 2017). Thus, Factor 2 might correspond to agricultural activities. Factor 3 was dominated by As, Pb, and Cd, with high loadings of 43.5%, 35.8%, and 35.3%, respectively. According to previous studies (Loska, and Wiechula 2003; Bhuiyan et al. 2015; Chen et al. 2019), industrial activities affect the distribution of As and Pb. Moreover, industrial activities are a primary source of Cd (Fang et al. 2019). Because there is no other industrial activity in the study area, the landfill is the principal industrial contributor. The thickness of the vadose zone (4–5 m) makes the transport of heavy metals from contaminated groundwater to the topsoil through capillary water difficult. Therefore, Factor 3 might represent a source dominated by landfill surface runoff. Factor 4 had higher loadings for Hg (36.8%) than for the other elements. Atmospheric deposition is a significant factor affecting the distribution of Hg (Gan et al. 2019). Due to its high volatility, Hg easily migrates to the topsoil through atmospheric deposition (Nannoni et al. 2019).
Additionally, heavy metal particles from vehicle exhaust on country highways and rural roads near the landfill site could be involved in dust deposition (Ozcan et al. 2007). Previous studies have shown that dust deposition increases heavy metal concentrations in the soil (Hernandez et al. 2003; Hu et al. 2020); furthermore, Qingdao has a temperate monsoon climate, which affects the deposition characteristics of dust (Li et al. 2016). Thus, Factor 4 may be associated with atmospheric deposition.

The heavy metals were attributed to natural, agricultural, landfill, and transportation sources (with contributions of 22.10%, 27.65%, 31.35%, and 18.89%, respectively) by combining the heavy metal spatial distributions, the Spearman correlation coefficients, and the PMF results. As the dominant contributor, the landfill affected heavy metal concentrations in the soil primarily through surface runoff.

Conclusions

In this study, the distribution characteristics, pollution evaluation, and source apportionment of heavy metals in soils around a landfill in Qingdao, China, were investigated for the first time. Based on the pollution assessment, heavy-metal contamination in soils in the area currently fall below thresholds for classification as polluted. Nevertheless, the health-risk assessment suggested that significant noncancer and cancer problems were likely to occur in adults based on the high levels of Cr and As, respectively. In addition, the comparison of background values and $I_{geo}$ for Cr, Cu, Ni, Zn, As, and Hg suggested the continuous accumulation of heavy metals in soils. Furthermore, Hg poses considerable potential ecological risk. The results indicated that the long-term negative effects of the landfill deserve extensive attention, despite the lack of a currently acute environmental impact. The source identification results indicated that the landfill contributed to a greater degree of heavy metal pollution (31.35%) than natural sources, agricultural activities, and transportation. Therefore, regular soil quality monitoring around landfills is suggested to prevent further deterioration related to heavy metal pollution. The surface runoff collection system should be strictly managed. This study provides a reference for effective landfill management strategies.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s00244-021-00857-9.

Author contributions Honghua Liu: Conceptualization, Methodology, Investigation, Data curation, Writing- Original draft preparation. Yuan Wang: Methodology, Investigation, Writing- Reviewing and Editing. Jie Dong: Investigation. Lixue Cao: Investigation. Lili Yu: Investigation. Jia Xin: Conceptualization, Supervision, Writing- Reviewing and Editing, Funding acquisition.

Funding This work was financially supported by the National Natural Science Foundation of China (41701619).

Availability of data and material The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Code availability Not applicable.

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