Distribution characteristics, pollution assessment, and source identification of heavy metals in soils around a landfill-farmland multi-source hybrid district

Honghua Liu
Qingdao Geo-Engineering Surveying Institute
Yuan Wang
Ocean University of China
Jie Dong
Qingdao Geo-Engineering Surveying Institute
Lixue Cao
Qingdao Geo-Engineering Surveying Institute
Lili Yu
Qingdao Geo-Engineering Surveying Institute
Jia Xin (xinj15@ouc.edu.cn)
Ocean University

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Honghua Liu [a], Yuan Wang [b], Jie Dong [a], Lixue Cao [a], Lili Yu [a], Jia Xin [b, *]

[a] Qingdao Geo-Engineering Surveying Institute (Qingdao Geological Exploration and Development Bureau); Key Laboratory of Urban Geology and Underground Space Resources; Shandong Provincial Bureau of Geology and Mineral Resources; Qingdao Geology and Geotechnical Engineering Co., Ltd, Qingdao, 266100, China

[b] Key Lab of Marine Environmental Science and Ecology, Ministry of Education; Shandong Provincial Key Laboratory of Marine Environment and Geological Engineering; College of Environmental Science and Engineering, Ocean University of China, Qingdao, 266100, China

Corresponding author*:

Dr. Jia Xin

College of Environmental Science and Engineering

Ocean University of China

Qingdao, 266100, China

Tel: +(86-0532) 66786568

E-mail address: xinj15@ouc.edu.cn

Abstract: Heavy metal pollution is one of the negative impacts deriving from municipal solid waste landfills. Due to 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, a combination of different pollution assessment methods and source identification tools are required to address pollution distribution and potential risks. In this study, the distribution 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. There are no environmental pollution and human carcinogenic risk posed by the heavy metals for the time being. However, high concentration of Cr in soil would cause non-carcinogenic risk for adults in the study area. Besides, the geo-accumulation indices of Cr, Cu, Ni, Zn, As, and Hg confirmed anthropogenic accumulation of these heavy metals in soils. Additionally, the potential ecological risk index indicated that Hg posed considerable risk to the eco-environment around the landfill. Sources of the heavy metals in the study area were attributed to natural sources (22.1%), agricultural activities (27.6%), and the landfill (50.3%). As the greatest contributor, the landfill affected heavy metal distribution in nearby surface soils mainly via surface runoff and waste gas. The continuous accumulation of heavy metals and non-carcinogenic risk for adults suggests the necessity for continuous monitoring of heavy metal content and migration around the landfill. This study provides a reference for local authorities in the study area.

Keywords: Soil; Heavy metals; Landfill; Distribution characteristics; Pollution

assessment.

1. Introduction

Heavy metals can exert negative effects when exceeding certain levels, although some of them are necessary elements for living beings and ecosystems (Huang et al. 2018). Due to their non-degradability, heavy metals remain in the host medium (Doumett et al. 2008; Adelopo et al. 2018). The majority (51.34-60.00%) of municipal solid waste in China is landfilled during 2015 to 2019 (China 2016, 2017, 2018, 2019, 2020), which have been defined as a critical source of heavy metal pollution. As highly stable contaminants, heavy metals could remain for up to 150 years in landfill sites with a leaching rate of 400 mm/year (EU 2002) and are easily transported to nearby soils (Adelopo et al. 2018). Thus, the evaluation of heavy metal pollution in soils around landfills and the associated risks is of vital importance.

A landfill site can impact the nearby soil environment through different pathways. First, heavy metals could migrate with surface runoff (Yusof et al. 2009), such as seeping into surrounding soils. Heavy metals in waste gases derived from landfills could also migrate into topsoil via atmospheric sedimentation (Nannoni et al. 2017). Finally, the leachate of municipal solid waste landfills can facilitate the adsorption of heavy metals on soil through vertical transport (Calace et al. 2001). Different dominant transfer pathways of heavy metals would result in varying distribution characteristics. Many studies have focused on the contamination and risks assessment posed by heavy metals in nearby soil near landfills. Different extents of heavy metal contamination were identified in soils around landfills in China, Turkey, Tunisia, Laos, Nigeria, and

Bangladesh (Zhou et al. 2015; Ogunbanjo et al. 2016; Vural et al. 2017; Vongdala et al. 2019; Alam et al. 2020; Wang et al. 2020). Heavy metals in soil, water, and plants near landfill was accumulated (Vongdala et al. 2019; Alam et al. 2020), which verified the potential environmental influence of landfill. Environmental management measures were suggested by researchers to reduce environmental risks in both non-polluting and polluting landfills (Zhou et al. 2015; Ogunbanjo 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 existed in landfills with longer running times (Chang et al. 2007). Due to its affinity for heavy metals, higher organic matter content might contribute to increased heavy metal leaching in landfills (Cao et al. 2013). Therefore, distribution characteristics and transport behaviors of heavy metals in soil around landfills have been investigated in many previous studies.

Usually, landfill sites do not stand alone, and are often located in the vicinity of 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 was associated with landfill leachate and agricultural activities (Liu et al. 2013b). However, most existing studies tended to assess heavy metal pollution in or around landfill site, without considering there might be potential impacts of other pollution sources (Bakis, and Tuncan 2011; Liu et al. 2013a). The positive matrix factorization (PMF) model can effectively realize source identification and apportionment, and has been applied widely 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 the source identification analysis.

A landfill in Qingdao, China, receiving 4300 tons of municipal solid waste daily, is suspected to pose a potential threat to the surrounding environment. In addition, there are villages and farmland around the landfill. Heavy metal distributions in the soil might thus be affected by multiple sources. Thus, it is necessary to evaluate the soil quantity around the landfill and identify the dominant pollution sources and pollution transport pathways. The aims of this study are: 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 distribution characteristics of these elements; and to identify sources and transport pathways of heavy metals in the study area.

2. Materials and methods

2.1 Study area

All samples were collected around a landfill located in Qingdao, Shandong Province, China. The landfill collects 4300 t/d of municipal solid waste from six main urban districts of Qingdao. The landfill was originally established for disposing waste in 2011. With increasing amount of waste, Xiaojianxi was expanded in 2017 by introducing

more environmental protection facilities and waste disposal facilities, such as some waste incinerators. The land-use types of the study area included farmland, woodland, residential area with livestock farms, and landfill, which accounted for about 50%, 10%, 10% and 30%, respectively. No industries apart from the landfill coexisted in the study area at the time of investigation. Qingdao is characterized by a mean temperature of 13.3 °C, annual precipitation of 722.7 mm, and the dominant wind direction is SSE, blowing particularly in spring and summer.

2.2 Sample collection and laboratory analysis

In June 2019, we collected 60 soil samples within a radius of 1 km away from the landfill. Because the southeastern part of the landfill is separated by a river, this area was not covered in our study. A map of the study area and sampling site distribution was illustrated in Fig. 1. To better represent variations in the spatial distributions of 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 group 1, 2, and 3, respectively.

The grid sampling method was used in the layout of the sampling sites. The sample (approximately 1.5 kg) for an individual sampling site was composed of 5 subsamples taken from 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, samples were sent for laboratory analysis.

The soil pH was determined by a pH meter (MAPRC 2006a), and the organic matter (OM) content was measured using oil bath-K_2CrO_7 titration (MAPRC 2006b). The total nitrogen (TN) content was measured by an automatic Kjeldahl nitrogen meter (AKNM; K1100F, Shandong Haineng Scientific Instrument, China). X-ray fluorescence spectrometry (XFP; Axios^{mAX}, Panak, Netherlands) was used to measure the concentrations of Ni, Cu, Pb, Zn, Cr, and total phosphorus (TP) (Lu et al. 2010; Kodom et al. 2012; Tang et al. 2015). Atomic fluorescence spectrometry (AFS; AFS-9750, Beijing Haiguang Instrument, China) was used for the determination of 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, NexlON 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 Qingdao Geo-Engineering Surveying Institute, which passed China Metrology Accreditation. To ensure quality control, reagent blank, duplicate samples, and certified reference standard soil samples (GBW07309, GBW07425 and GBW07451) were included in the analytic process. The relative deviation (RD) should be controlled 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.

2.3 Assessment of heavy metal contamination

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

The single pollution index (PI) refers to the pollution level of each heavy metal in the soil, while the Nemerow integrated pollution index (NIPI) indicates the total heavy metal pollution level of the study area. The values of PI for individual elements (PI_i) and NIPI were calculated according to Eqs. (2) and (3), respectively:

$$PI_i = C_i / C_{b,i} \tag{2}$$

$$NIPI = \sqrt{\frac{\left(\overline{PI_i}\right)^2 + \left(PI_{\max}\right)^2}{2}} \tag{3}$$

where C_i indicates the content of each individual element (mg kg^{-1}), $C_{b,i}$ indicates its corresponding value specified in the Soil environment quality Risk control standard for soil contamination of agricultural land (mg kg^{-1}) (MEEPRC 2018), $\overline{PI_1}$ indicates the mean PI value (unitless), and PI_{max} indicates the maximum PI value (unitless). The classifying criteria for PI and NIPI are summarized in Table S1 (Nemerow 1985; Fei et al. 2019).

The geo-accumulation index (I_{geo}), originally used for sediments, is a widely used assessment method to reveal the enrichment of elements in soil (Loska et al. 2004). It can be calculated using Eq. (4):

$$I_{geo} = \log_2 \frac{C_i}{1.5C_{b,i}} \tag{4}$$

where C_i represents the content of the individual metal element (mg kg^{-1}) and $C_{b,i}$ represents its 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, Hg=0.035) (Zhang 2011). As shown in Table S2, I_{geo} can be divided into 7 levels (Muller 1969).

Potential ecological risk is usually applied in the estimation of the effects that heavy metals may exert on an ecological system. The ecological risk index (ER) of each element is calculated according to Eq. (5), and the overall potential ecological risk index (PER) is given as the sum of all ERs:

$$ER_i = T_i \times \frac{C_i}{C_{b,i}} \tag{5}$$

$$PER = \sum_{i=1}^{n} ER_i \tag{6}$$

where C_i indicates the content of individual metal elements (mg kg^{-1}), $C_{b,i}$ indicates its 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, Hg=0.035) (Zhang 2011), and T_i indicates the toxicity coefficient of each metal element (unitless, where the values are As=10, Cd=30, Cr=2, Cu= Ni=5, Hg=40, Pb= Zn=1) (Hakanson 1980). According to its value, the potential ecological risk index can be classified into several levels (Table S3).

2.4 Health risk assessment

Health risks for both adults and children were estimated in the study area. Exposure pathways of metal elements mainly comprised ingestion, inhalation, and dermal contact, which are herein referred to as D_{ing}, D_{inh}, and D_{dermal} (mg kg^{-1} d^{-1}), respectively. The main ingestion pathway is the consumption of vegetables in the study area. The concentrations of heavy metals in vegetable and mean daily doses received through the individual exposure pathways were calculated using Eqs. (7), (8), (9), and (10) (USEPA 1989; Wang et al. 2005; USEPA 2015):

$$C_{vegetable} = C_{soil} \times TF_i \tag{7}$$

$$D_{ing} = C_{vegetable} \times \frac{IngR_{vegetable} \times EF \times ED}{BW \times AT} \tag{8}$$

$$D_{dermal} = C_{soil} \times \frac{SL \times CF \times SA \times ABS \times EF \times ED}{BW \times AT} \tag{9}$$

$$D_{inh} = C_{soil} \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \tag{10}$$

where $C_{vegetable}$ represents the content of the individual metal element in vegetable (mg kg^{-1}), C_{soil} represents the content of the individual metal element in soil (mg kg^{-1}), TF_i represents the ratio of element contents in vegetables to soil (unitless), $IngR_{vegetable}$ represents the ingestion rate of vegetable for people (mg d^{-1}), CF represents the conversion factor applied in mathematical calculations (kg mg^{-1}), FI represents the fraction of intake (unitless), EF represents the exposure frequency of people to the pollutant (day $year^{-1}$), ED represents the duration of exposure of people to the pollutant (year), BW represents the mean body weight of people (kg), AT represents the mean exposure time for people (days), SL represents the skin adherence factor for people (mg cm^{-2} h^{-1}), SA represents the mean skin area of people (cm^2), ABS represents the dermal absorption factor of people (unitless), InhR represents the inhalation rate of soil (m^3 d^{-1}), and PEF represents the particle emission factor (mg^3 kg^{-1}). The values of these parameters are summarized in Table S4 (USEPA 2001; Zheng et al. 2010; Wang et al. 2005) and Table S5 (Bian et al. 2014; Hu et al. 2014).

The non-carcinogenic risk was estimated by Eq. (11), and the carcinogenic risk was calculated with Eq. (12) (USEPA 1989):

$$HI = \sum HQ_i = \sum \frac{D_i}{RfD_i} \tag{11}$$

$$Risk(RI) = \sum D_i \times SF_i \tag{12}$$

where RfD_i indicates the reference dose for a single heavy metal (mg kg^{-1} d^{-1}) and SF_i indicates the carcinogenic slope factor for a single heavy metal (mg kg^{-1} d^{-1}). The values of these parameters are summarized in Table S5 (Xiao et al. 2015; Pan et al. 2016; Eziz et al. 2018).

2.5 Source identification

The PMF model is recommended by the United States Environmental Protection Agency (USEPA) for source identification (Norris et al. 2014). As shown in Eq. (13), 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):

$$X_{ij} = \sum_{k=1}^{p} G_{ik} F_{jk} + E_{ij} \tag{13}$$

where subscripts i, j, and p represent the amount of samples, elements, and factors, respectively. After minimizing the objective function Q, E_{ij} is obtained:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{E_{ij}}{\mu_{ij}} \right)^2 \tag{14}$$

where μ_{ij} represents the uncertainty of concentration values. In this study, uncertainties were calculated following Eq. (15):

$$\mu_{ij} = 0.1 x_{ij} + \frac{MDL}{3} \tag{15}$$

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

Two input files PMF model required were the concentrations of samples and the uncertainty of sample data. Then, output results from PMF could aid source identification of pollutants.

2.6 Data analysis

Spearman correlation coefficients were applied in the analysis of data under non-normal distribution. Microsoft Excel version 2016 and SPSS version 25 were used for statistical processing. The spatial distributions of metal elements were obtained by ArcGIS software version 10.2, using inverse distance weighted (IDW) methods. PMF version 5.0 was used to identify potential pollution sources.

3. Results and discussion

3.1 Soil properties

The statistical data for soil properties including pH value, OM, TN, and TP of soil around the landfill are shown in Table S6. It was inferred that the soil was acidic (pH=5.77), which could increase the release of heavy metals (Chai et al. 2007). In comparison to the background (CNEMC 1990), soil samples in the study area contained higher contents of OM (2.15%) and TN (1.23 g kg^{-1}). The background value of TP (0.66 g kg^{-1}) (CNEMC 1990) was similar to the average TP content in soil samples.

3.2 Concentrations of heavy metals

Table 1 illustrates the statistical data of metal element concentrations in soils around the landfill site studied, and its comparison with concentrations in 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. As listed in

Table 1, the heavy metals in this study would not cause soil contamination, with concentrations lower than the risk screening values of agricultural land in China (MEEPRC 2018). However, the mean contents of most metal elements (Cr, Cu, Ni, Zn, As, and Hg) were beyond their corresponding background contents in Qingdao (Zhang 2011). Moreover, the maximum concentrations of As and Hg were approximately three times the corresponding background values, indicating heavy accumulation of these elements. It was therefore inferred that these heavy metals were accumulated due to anthropogenic activities. 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).

It can be seen in Table 1 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 landfill studied (4300 t/d) were significantly higher than those near a landfill in Vientiane, Laos (300 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 landfill studied (8 years in operation) contained lower concentrations of Cr, Cu, and Ni than soils around a municipal solid waste landfill in Pilsen, Czech (21 years in operation; 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; 86.00 and 34.00 mg kg^{-1} for Cr and Ni, respectively) (Liu et al. 2013b), and the Ginestreto municipal solid waste landfill in Italy (22 years in operation; 89.00, 25.50, and 54.80 mg kg^{-1}, respectively) (Nannoni et al. 2017), which might be a result of the shorter time in operation. 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. However, the landfill under study contained higher concentrations of Pb and As than the three landfill examples with longer operating times, which might be associated with the specific waste composition (Ishchenko 2019).

3.3 Heavy metal contamination assessment

3.3.1 Pollution index

Table S7 illustrates the assessment results including pollution classes for the topsoil samples. The PI values for eight of the metal elements studied were less than 1. This demonstrated that eight heavy metals for all samples were under the safe class, showing little threat to the quality and safety of agricultural production. In addition, NIPI value (0.40) also suggested there was no environmental pollution caused by heavy metals.

3.3.2 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 summarizes the pollution level classification according to I_{geo} evaluation results. The percentages of samples which ranged from uncontaminated to moderately contaminated class are 31.67%, 20.00%, 38.33%, 1.67%, 81.67%, and 33.33%, respectively, for Cr, Cu, Ni, Zn, As and Hg, showing varying degrees of accumulation for different heavy metals in soil. It could be inferred that the accumulation of different heavy metals in soil might be attributed to different affecting factors. With 81.67% of the samples found contaminated with As, it was indicated that

As was heavily enriched in soils around the landfill, which was affected by unnatural factors seriously.

3.3.3 Potential ecological index

In this study, ER and PER were calculated to estimate ecological risk, and are illustrated in Table S9. 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 samples posed considerable risk in terms of Hg content. Thus, Hg should be considered a priority pollutant among the eight elements studied. According to the categorization standard shown in Table S3, the mean PER value (117.21) indicated that the ecological environment near the landfill site was under low risk of heavy metal pollution.

3.4 Health risk assessment

Table 2 summarizes the risks posed to human health by the eight heavy metals in soil via various exposure pathways. Overall, for most heavy metals, they followed the similar order in terms of risk level regardless for adults or children. However, there was significant difference between the risk level of Cr for adults and children. Besides 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^{-1}, 1.05×10^{-1}, 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). For adults, Cr

held the highest HI value, whereas Cr held the second lowest HI value for children. In addition, the HI value for Cr for adults (5.57) was higher than children (1.31×10^{-1}). The reason might be that the concentration of Cr in soil was relatively high, and adults (0.345 kg d^{-1}) generally took more vegetables than children (0.2315 kg d^{-1}). Ingestion, inhalation, and dermal contact were the exposure pathways to the heavy metals contained in soils. The hazard quotient (HQ) for each pathway decreased in the order of $HQ_{ing} > HQ_{dermal} > HQ_{inh}$. Thus, ingestion was the dominant exposure pathway to heavy metals in this study. With HI value for adult higher than 1, the exposure to heavy metals would cause non-cancer-related health problems for adults in the study area, which must be paid attention seriously.

The carcinogenic risks were estimated for certain heavy metals (Cr, Ni, As, and Cd) by the SF value. The carcinogenic risks posed to both populations (adults and children) followed the same order, of Cr > As > Ni > Cd. The cancer risks posed by Cr, Ni, As, and Cd to adults were 2.56×10^{-7}, 2.09×10^{-9}, 1.46×10^{-8}, and 5.66×10^{-11}, respectively, which were higher than those posed to children (1.57×10^{-7}, 1.28×10^{-9}, 8.97×10^{-9}, and 3.47×10^{-11}, respectively). These results corroborate those reported in a previous study that also stated that adults are subjected to greater cancer risks than children (Yang et al. 2018). Overall, the carcinogenic risks to humans were lower than 1.00×10^{-6}, indicating that significant health problems are not expected to arise (Fryer et al. 2006).

3.5 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 soils of the study area.

Table 3 summarizes the comparison between the three groups of sampling sites based on multiple indexes including C_{soil}, PI, NIPI, I_{geo}, 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, the health risks posed by Cu, Pb, As, and Hg followed the same order. Moreover, the I_{geo} 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. The distribution maps of metal elements estimated with IDW methods in GIS technology are shown in Fig. 2. Hotspots in spatial distribution usually represent high levels of concentration and pollution (Doyi et al. 2018). It can be inferred from Fig. 2 that there were two hotspots located in the northwestern (group 2) and southwestern (group 3) areas around the landfill, which is consistent with the results presented above. Since a hotspot in the northwestern area was located in an impact area of the prevailing wind, the distribution of heavy metals might be associated with the prevailing wind, as was reported in a previous study (Li et al. 2017). Due to more exposure to atmospheric deposition, heavy metal concentrations tend to be higher at higher altitudes (Cao et al. 2013). However, the hotspot in the southwestern area was at low elevation. Surface runoff was suspected to be another dominant transport pathway of heavy metals from the landfill. Additionally, hotspots corresponded roughly to the positions of two villages

wherein various agricultural activities are realized. Thus, the overall heavy metal contamination may be from a combination of landfill and agricultural activity.

Spearman correlation coefficients were used to investigate the correlations among the eight heavy metals. As listed in Table S11, Cr was remarkably positively correlated with Ni (r=0.955). Additionally, Cu exhibited a strong positive correlation with Zn (r=0.919). The two elements in these pairs may thus originate from similar sources.

The PMF model revealed more supportive evidence for the identification and apportionment of the potential sources of heavy metals. Concentration data for 60 samples along with uncertainties were inputted into the model. Based on the minimum and most stable value of Q, there should be 4 factors, and the result is shown in Fig. 3. Factor 1 was ruled by Cr and Ni with high factor loadings (33.1% and 41.5%, respectively), which corresponds with the results of distribution characteristics and the Spearman correlations, further suggesting the same source for Cr and Ni. It was previously reported that the distribution of Cr and Ni is dominated by natural sources such as parent rocks (Lu et al. 2012; Ke et al. 2017; Zhang et al. 2018). Thus, it was proposed that factor 1 is related to a natural source. Factor 2 was dominated by Cu and Zn with loadings of 35.6% and 39.1%, respectively. According to our survey, there were private and centralized livestock farms nearby, where wastewater and solid wastes generated by farming activities are discharged directly into the soil, causing the accumulation of Cu and Zn (Cang et al. 2004). Moreover, pesticides and fertilizers are commonly used in the farmland within the study area, and excessive application of phosphate fertilizers would lead to 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, presenting 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 represent a primary source of Cd (Fang et al. 2019). As there is no other industrial activity in the study area, the landfill is the main industrial contributor. Considering the vadose zone thickness (4–5 m), it would be difficult for heavy metals to be transported from contaminated groundwater to the topsoil through capillary water. Therefore, factor 3 might represent a source dominated by landfill surface runoff. Factor 4 had higher loadings for Hg (36.8%) than for 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 topsoil through atmospheric deposition (Nannoni et al. 2017; Lv 2019). Thus, factor 4 may be associated with atmospheric deposition of landfill gas.

Combining the heavy metal spatial distributions, Spearman correlation coefficients, and PMF results, the heavy metals were attributed to three sources, namely natural sources, agricultural activities, and the landfill, with contributions of 22.1%, 27.6%, and 50.3%, respectively. As the dominant contributor, the landfill affected heavy metal concentrations in soil mainly through surface runoff and waste gas deposition.

4. 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, there was no heavy metal contamination at the time of investigation. Nevertheless, the health risk assessment suggested that significant non-cancer problems were likely to occur for adults based on the high level of Cr present. Besides, the comparison to background values and the I_{geo} for Cr, Cu, Ni, Zn, As, and Hg suggested that the accumulation of heavy metals in soils was occurring continuously. Furthermore, Hg was found to pose considerate potential ecological risk. The results indicated that the long-term negative effects of the landfill deserve broad attention, even though its acute environmental impact was not serious at the time of study. The source identification results reflected that the landfill contributed to heavy metal pollution to a greater degree (50.3%) than the natural sources or agricultural activities. Surface runoff and waste gas were the main transport pathways causing contamination. Therefore, regular soil quality monitoring around the landfill is suggested for preventing further deterioration related to heavy metal pollution. The surface runoff collection system and the waste gas treatment system should be further strictly managed. This study further provides a reference for effective landfill management strategies.

5 Declarations

5.1 Funding

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

5.2 Conflicts of 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.

5.3 Availability of data and material

The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

5.4 Code availability

Not applicable

5.5 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.

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Figures

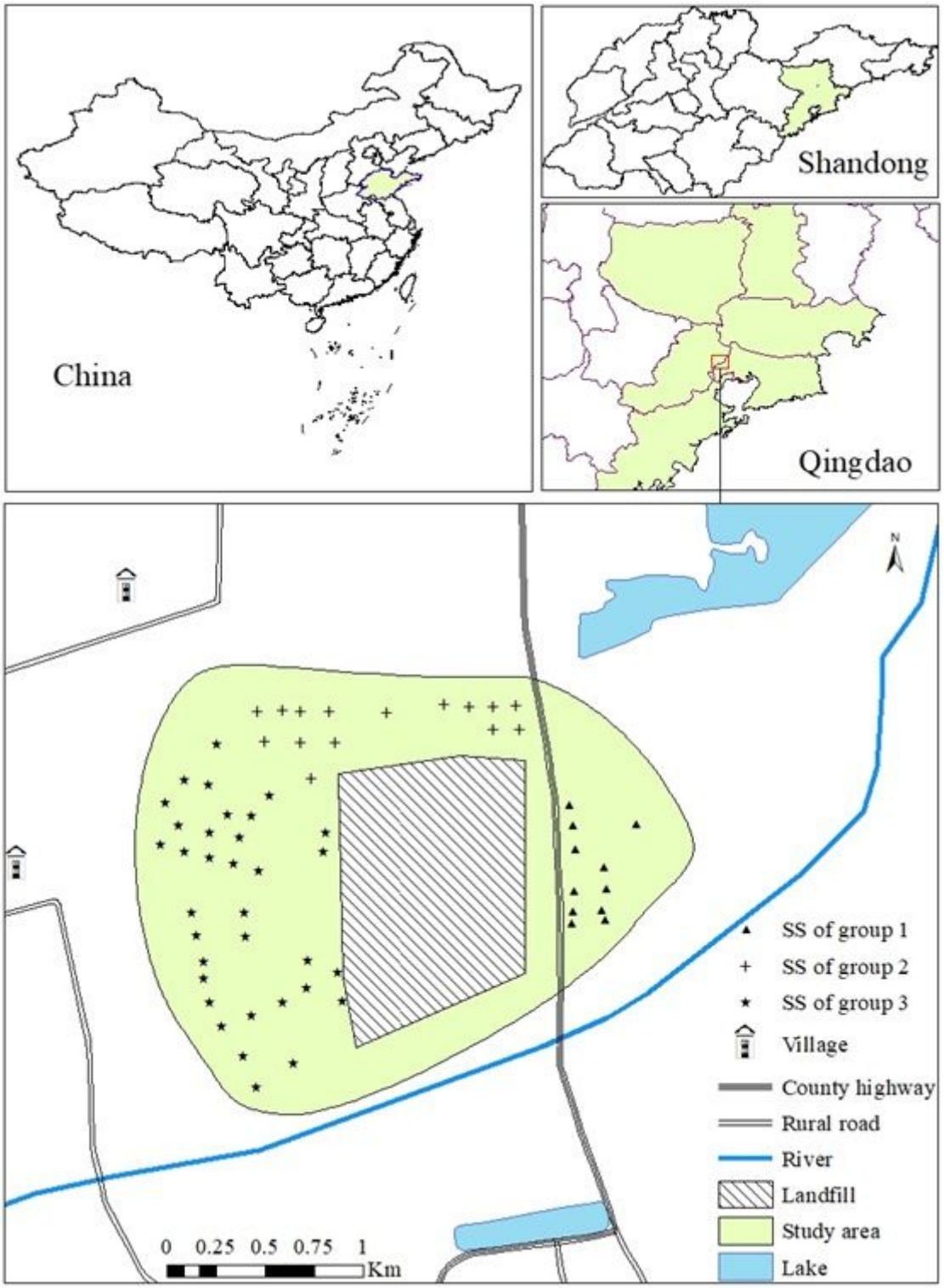

Figure 1

Location of the study area and distribution of sampling sites (SS). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its

authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

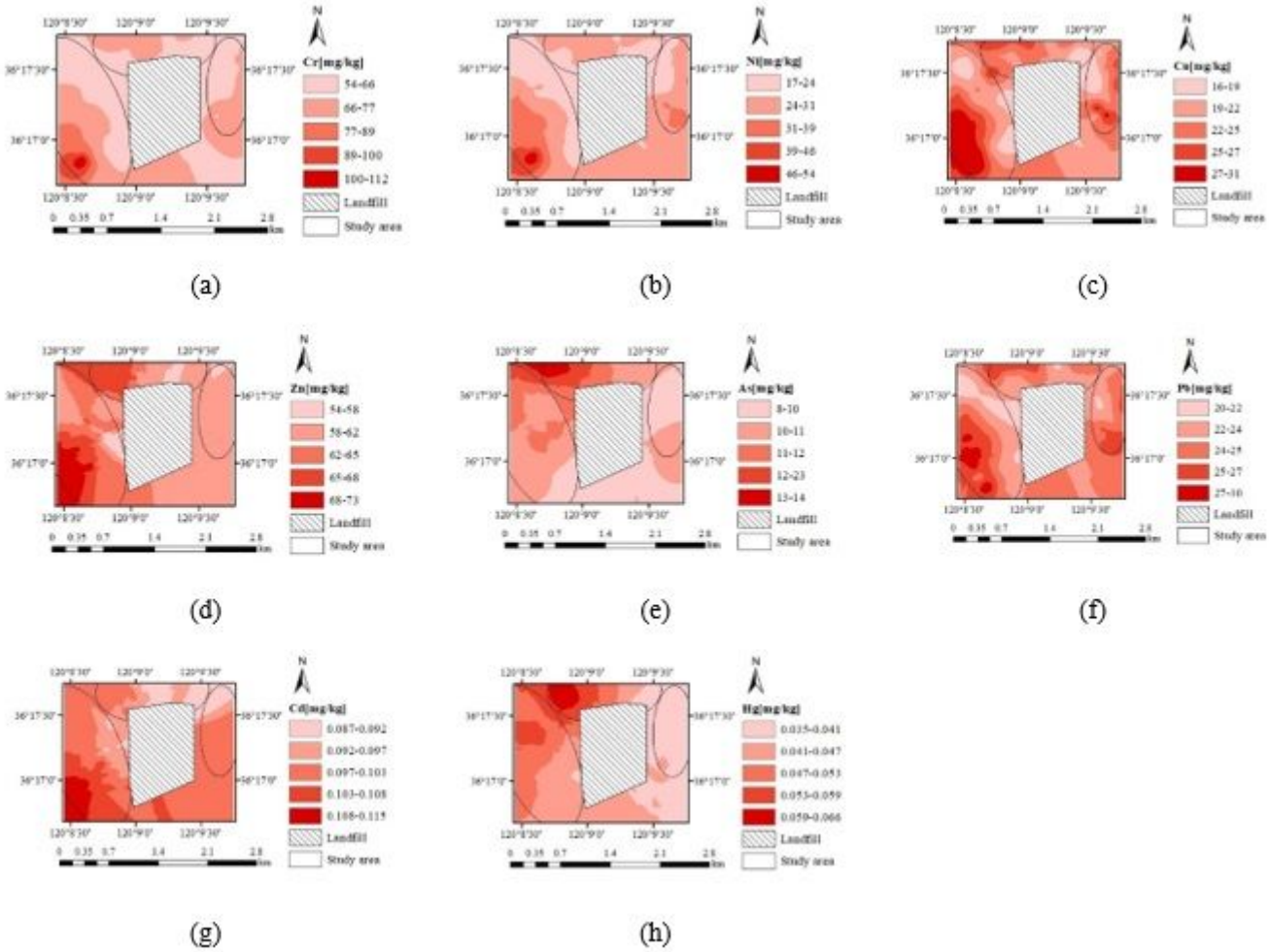

Figure 2

Spatial distributions of the concentrations of: (a) Cr; (b) Ni; (c) Cu; (d) Zn; (e) As; (f) Pb; (g) Cd; (h) Hg.

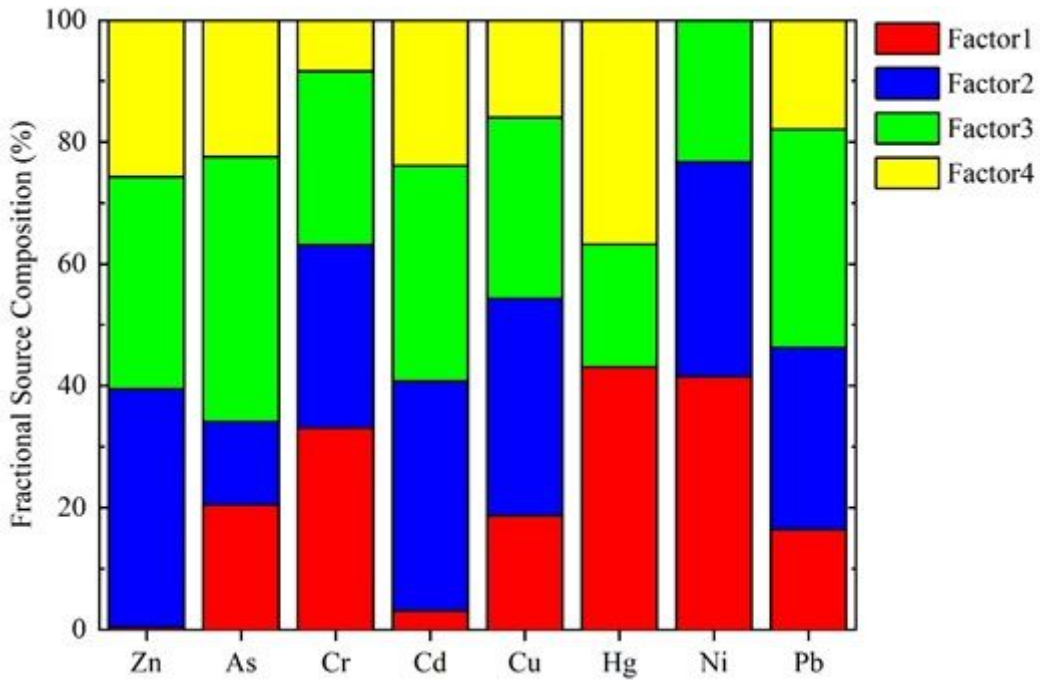

Figure 3

Factor profiles of heavy metals in soils around the landfill, derived from the PMF model.

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