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Risk assessment and source identification of heavy metals in agricultural soil: a case study in the coastal city of Zhejiang Province, China

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Abstract

Heavy metal contamination is a serious environmental problem, especially in developing countries such as China. In this study, we collected 1928 soil samples from the southeastern coastal area of China and analyzed the pollution concentration and potential ecological risk from heavy metals including arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and mercury (Hg). The mean concentrations of Cr, Hg, and Pb were lower than their corresponding background values, whereas As and Cd were 1.31 and 1.59 times their background values, respectively. The calculation of the mean Pollution Index (*PI*) for these heavy metals were, in decreasing order Cd (1.59), As (1.31), Cr (0.94), Pb (0.89), and Hg (0.78) and the Nemerow Integrated Pollution Index revealed that almost one-fifth of the soil in the study area was moderately polluted. According to the ecological risk index, about 12% of the soil was at a moderate or high ecological risk, and Cd and Hg presented the highest ecological risk. The GeogDetector software was used to quantitatively assess the potential sources of these metals. The GeogDetector results showed that the soil heavy metals have various sources, including: natural processes had significant impacts on all heavy metals analyzed in this study; farmland types influenced the concentrations of As and Cr significantly; industrial activities significantly increased As, Cr, and Hg; transportation-related activities increased As, Cd, and Hg; and agricultural application of fertilizer and pesticides, had significant impacts on As, Cd, and Pb levels. Based on the results of the interaction detector, natural processes and agricultural activities were determined to be the main sources of heavy metals in the study area.

Keywords Heavy metal · Source analysis · Pollution assessment · GeogDetector model

1 Introduction

Heavy metal contamination in soil has become an important global environmental concern in recent decades and has thus received significant attention (Chen et al. 2009; Islam et al. 2016; Jiang et al. 2017a). As a developing country with rapid urbanization and industrialization, heavy metal pollution is a serious environmental hazard in China, especially in areas around industrial zones (Yu et al. 2012; Yang et al. 2017; Liu et al. 2019). Heavy metals usually have long residence times and persistent bioavailability, and can accumulate in agricultural soils, leading to gradual decreases in soil fertility, degeneration of soil biology, and reduction in crop productivity (Nanos et al. 2015; Xu et al. 2017; Zhao et al. 2014). Moreover, they easily enter plants, animals, and humans by inhalation, dermal absorption, or ingestion, and are thus biomagnified

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through the food chain, posing a grave risk to food safety and human health (Sun et al. 2010; Zang et al. 2017). Hence, there is an urgent need to quantitatively assess the characteristics of soil heavy metal pollution and identify the sources of heavy metals, to address the threats to soil quality, food safety, and human health.

In recent years, several studies on soil heavy metal pollution have been conducted in China, especially on pollution assessment and source analysis (Jiang et al. 2017a; Marrugo-Negrete et al. 2017; Zhao et al. 2014). Correspondingly, various indices have been widely used for heavy metal pollution assessment in soil, such as Pollution Index, Geoaccumulation Index, Contamination Factor, Enrichment Factor, Nemerow Integrated Pollution Index and Sediment Pollution Index (Hu et al. 2017; Loska et al. 2004; Nemerow 1985). Furthermore, other indices have been implemented to assess the impact of heavy metal pollution on environment or human health, such as the Potential Ecological Risk Index (Hakanson 1980), Hazard Index and Carcinogens Risk Factors (USEPA 1997). Generally, heavy metals are present in soil due to natural processes and human activities. Natural sources of heavy metals are mainly controlled by their parent materials, while anthropogenic sources include atmospheric deposition, fertilizers and pesticides, mining, coal combustion, transportation, metalliferous industries, etc. (Liu et al. 2015; Zhang 2006). Various multivariate statistical methods, such as principal component analysis, cluster detection, multiple linear regression, and positive matrix factorization have been widely used for complex dataset interpretation and source identification (Luo et al. 2015; Qiao et al. 2011; Ming-Kai et al. 2013). However, most studies focus on qualitative source identification, and little is known about quantitative source apportionment. Moreover, existing studies have generally classified heavy metals according to a single source (natural or anthropogenic), and a multi-source analysis of heavy metals is thus lacking. Therefore, in this study, we conduct an intensive survey to quantitatively assess the pollution risk of heavy metals, and identify the multi-sources of heavy metals in agricultural soils using a novel geographical detector model (Wang et al. 2010).

The aims of this paper are as follows: (a) determine the concentrations of heavy metals (As, Cd, Cr, Hg, and Pb) in agricultural soils, (b) assess the pollution levels and potential ecological risks of these heavy metals, and (c) quantitatively calculate the contribution of each source to heavy metal pollution. The results of this study can potentially provide valuable information for soil quality control and management, food safety insurance, and human health protection.

2 Materials and methods

2.1 Study area

The area of study, located in the southeastern coastal area of China (29°11′–30°33′N, 118°21′–120°30′E) is one of the most important cities in the Yangtze River Delta city group and the Ninghang ecological economic zone. It has a subtropical monsoon climate, with hot and humid summers, and cold and dry winters. According to statistical yearbook of the study area (http://tjj.hangzhou.gov.cn), the annual average temperature, average humidity, precipitation, and sunshine hours are 17.8 °C, 70.3%, 1454 mm, and 1765 h. respectively. The southwestern region of the study area is part of the western hilly areas of the Zhejiang Province, while the northeastern region is part of the northern plain area of Zhejiang. The main soil types are red soil and paddy soil derived from eluvium and alluvial soil parent materials respectively. The area of study covers 16,596 km^2, and its total population was about 9.19 million at the end of 2015 (http://tjj.hangzhou.gov.cn). Over the past three decades, rapid urbanization and industrialization have caused serious environmental problems in this city, including heavy metal pollution from anthropogenic sources (Zhang and Wang 2009). Chemicals, machinery, nonmetallic minerals, metallurgy, and papermaking are the top five industries (Fei et al. 2018a). As one of the most important food production bases in Hang-jia-hu Plain, heavy metal pollution in agricultural soil has profound impacts on local food safety and human health. However, few studies have been able to conduct high density investigate of soil pollution in agricultural soil and evaluate the pollution sources in the context of spatial heterogeneity.

2.2 Data sources

A total of 1928 samples of topsoil (0–15 cm) were taken from the centers of 1 km grid squares in 2015 (Fig. 1a). All the in situ samples were from rice, fruit, vegetable, and tea farms. Since this study focuses on agricultural soil, the downtown area was not sampled due to a lack of agricultural soil. Five sub-samples around each sampling point were collected and mixed thoroughly to get a representative sample. With the help of the Global Positioning System (GPS) the location of each sample site was recorded. All soil samples were packed into polyethylene bags and brought back to the lab, then air-dried at room temperature and ground through 100 meshes for chemical analysis. To analyze arsenic (As), cadmium (Cd), chromium (Cr), and lead (Pb), the soil samples (0.5 g) were acid-digested by a $HCl–HNO_3–HClO_4$ mixture and their concentrations were determined by plasma mass spectrometry (ICP-MS, TMO,

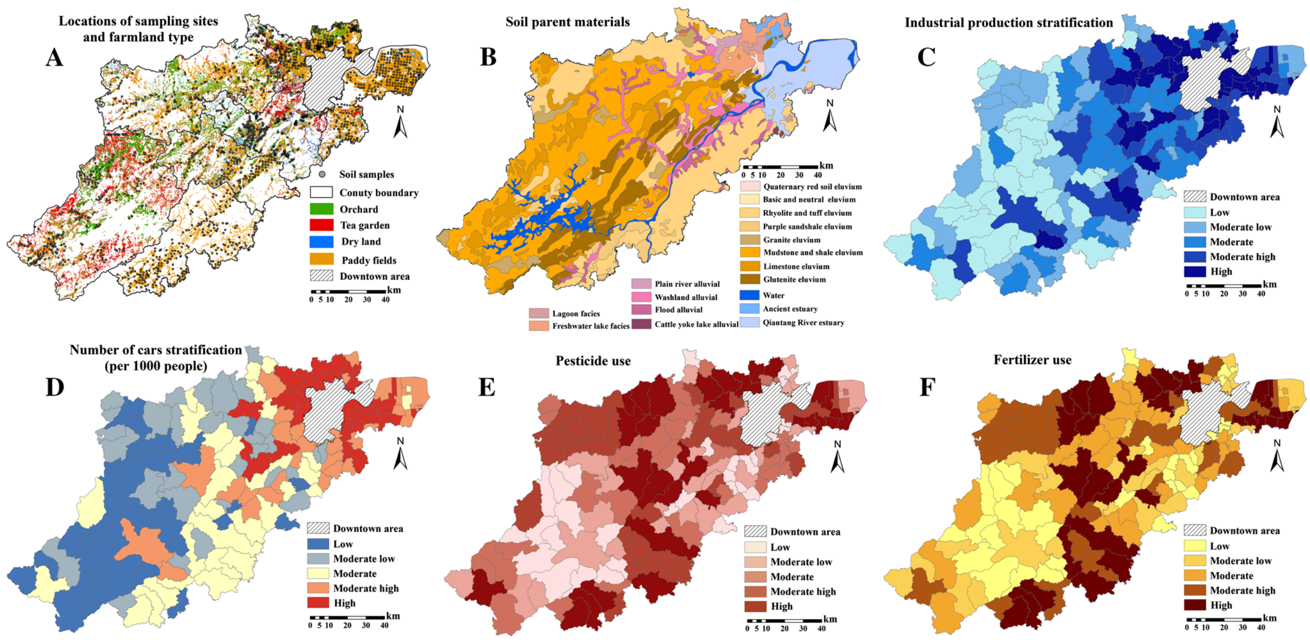

Fig. 1 The distribution of soil samples and contribution factors (sources) (**a** sampling sites and farmland type, **b** soil parent materials, **c** industrial production, **d** Number of cars per 1000 people, **e** pesticide use, **f** fertilizer use)

USA) (Yang et al. 2017). For mercury (Hg), the soil samples were digested by a mixture of nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) in a microwave-accelerated reaction system, and its concentration was determined by atomic fluorescence spectrometry. Blind duplicates and standard reference materials (GSS-3, China National Center for Standard Materials) were used for quality assurance and control. Standard sample recovery ranged between 90 and 110%, and the relative standard deviations of the duplicate samples were between 3 and 8% (Fei et al. 2018b).

Previous studies have proven that natural sources also contribute to the concentrations of heavy metals in soils (Wang et al. 2015; Liu et al. 2017). To quantitatively assess their influence, the distribution of soil parent materials (Fig. 1b) was obtained from the soil database of Zhejiang Province (Wu et al. 2013). Furthermore, to quantitatively assess the impact of various anthropogenic sources on the distribution of heavy metals (Liu et al. 2015; Zhang 2006), data of industrial production in terms of ten thousand yuan (proxy for industrial activities), number of cars per thousand people (proxy for traffic), and pesticide and fertilizer use in tons (proxy for agricultural activities) were obtained from the Hangzhou Statistical Yearbook in 2015, and their distribution is shown in Fig. 1c–f. As previously stated, the downtown area was not sampled and consequently, data of the anthropogenic sources in the downtown area was not collected.

2.3 Heavy metal pollution assessment

The pollution index was used to assess the degree of contamination of each heavy metal, and is calculated as follows (Zang et al. 2017):

$$PI = \frac{C_i}{C_{b,i}} \tag{1}$$

where PI is the pollution index, C_i is the concentration of the ith heavy metal in the soil, and $C_{b,i}$ is its corresponding background value in the study area (Xu et al. 2012). Based on the PI values, five categories were defined: unpolluted ($PI \leq 1$), slightly polluted ($1 < PI \leq 2$), mildly polluted ($2 < PI \leq 3$), moderately polluted ($3 < PI \leq 5$), and highly polluted ($PI > 5$) (Zang et al. 2017). Since PI only represents the contamination of single heavy metal, the Nemerow Integrated Pollution Index ($NIPI$) (Nemerow 1985) was used to assess the overall heavy metal pollution status of the soil. $NIPI$ is calculated as follows:

$$NIPI = \sqrt{\frac{(P_{\max})^2 + (\bar{P}_i)^2}{2}} \tag{2}$$

where $P_{\max}$ is the maximum value of all pollution indices (PIs as calculated above) of the soil heavy metals, and $\bar{P}_i$ is the average value of the pollution indices. Five categories were defined according to the $NIPI$: no pollution ($NIPI \leq 0.7$), pollution warning threshold ($0.7 < NIPI \leq 1$), low pollution ($1 < NIPI \leq 2$), moderate pollution ($2 < NIPI \leq 3$), and severe pollution ($NIPI > 3$) (Nemerow 1985).

Considering the toxicology of heavy metals, the ecological risk index (*RI*) was also used to assess the ecological risk posed by heavy metals in soils (Hakanson 1980), and was estimated as follows:

$$RI = \sum_{i=1}^{n} ER_i \tag{3}$$

$$ER_i = T_i \times \left(\frac{C_i}{C_{b,i}}\right) \tag{4}$$

where ER_i is the ecological risk index for the heavy metal *i*, and T_i is the toxicity response coefficient for the metal *i*. The toxic-response factors for As, Cd, Cr, Hg, and Pb are 10, 30, 2, 40, and 5 respectively (Wang et al. 2015). C_i is the concentration of the heavy metal *i* in the soil, and $C_{b,i}$ is its corresponding background value (Xu et al. 2012). *ER* indicating the single ecological risk of each heavy metal is defined as five categories: low risk ($ER < 40$), moderate risk ($40 \le ER < 80$), considerable risk ($80 \le ER < 160$), high risk ($160 \le ER < 320$), and very high risk ($ER \ge 320$). *RI* representing the total ecological risk of evaluated heavy metals is defined as four categories: low risk ($RI < 150$), moderate risk ($150 \le RI < 300$), considerable risk ($300 \le RI < 600$), and high risk ($RI \ge 600$) (Hakanson 1980).

2.4 Statistical analysis

In this study, GeogDetector—a novel relative spatial variance analysis tool that works for both numerical and categorical variables—was used to quantitatively assess the contribution factors (sources) of heavy metal contamination (Wang et al. 2010). The basic assumption of GeogDetector is that when a heavy metal contaminant is present and a contribution factor (source) can be determined for that particular heavy metal, its concentration in the area exhibits a spatial distribution similar to that of the contribution factor (Wang and Hu 2012). The process using GeogDetector is shown as a flowchart in Fig. 2. First, the locations of heavy metals were converted to grid points i_1, i_2, …, i_n though ordinary kriging technique (Fei et al. 2019, Olea 2006) in Arcmap 9.3 software. Then, contribution factors such as pesticide or fertilizer use (layers *S*, *E*) were classified into different sub-regions (s_1, s_2, $s_{3\ldots}$ and e_1, e_2, $e_{3\ldots}$) according to the principle of minimizing the dispersion variance within each sub-region and maximizing the dispersion variance between each sub-region. Subsequently, the heavy metal layer *I* was overlaid with the contribution factor layers (*S* or *E*) and the area of each sub-region and the corresponding variance of heavy metal concentration in each sub-region was then calculated. Finally, to quantitatively assess the contribution of each factor (source) to heavy metal concentration, the *q* value was estimated as follows (Wang et al. 2010):

$$q = 1 - \frac{1}{N\sigma^2}\sum_{i=1}^{n} N_i \sigma_i^2 \tag{5}$$

where *N* is the total area of the study region, σ^2 is the overall variance of heavy metal concentration, N_i and σ_i^2 are the area and heavy metal variance of the sub-region *i*, respectively, and *n* is the total number of sub-regions. The *q* value measures the impact strength (contribution), such that a *q* value ranging from 0 to 1 represents the impact (from the weakest to the strongest) of a given contribution factor on the heavy metal concentration (Wang et al. 2010). If the heavy metal (*I*) is completely controlled by a given contribution factor (*S*), the variance of its concentration in every sub-region should be 0, thus, $q = 1$. Whereas if the heavy metal (*I*) is totally uncorrelated to the contribution factor (*S*), we get $q = 0$. Additionally, using this model, the significance of the contribution of each source, the difference in average heavy metal concentrations between sub-regions, and the interaction between the contributions of two sources can be quantitatively calculated (Li et al. 2013, Fei et al. 2016). The relevant calculations were made using the GeogDetector software (Wang and Hu 2012) (www.sssampling.org/geogdetector).

3 Results and discussions

3.1 Descriptive statistics of heavy metal concentrations in agricultural soils

The descriptive statistics of heavy metal concentrations in the agricultural soils of the study area, the background concentrations in Zhejiang province, and the risk screening values for each heavy metal as defined in the Soil environmental quality in China (GB 15618-2018), are shown in Table 1. If the concentration of a heavy metal in soil exceeds its corresponding risk value, it is considered harmful to human health (Liang et al. 2017). The concentrations of As, Cd, Cr, Hg, and Pb ranged from 2.63 to 43.20, 0.05 to 1.42, 10.50 to 104.00, 0.03 to 0.50, and 17.00 to 62.60 mg/kg with the median values of 7.07, 0.20, 54.40, 0.11, and 30.90 mg/kg, respectively. The mean concentrations of As, Cd, Cr, Hg, and Pb were 8.99, 0.27, 52.90, 0.13, and 31.66 mg/kg, respectively, all of which are lower than the Environmental Quality Standard risk screening values. However, compared to their local background values, the mean concentrations of Cr, Hg, and Pb were lower, whereas the mean concentrations of As and Cd were higher. The mean concentrations of As and Cd were 1.31 and 1.59 times their respective background values.

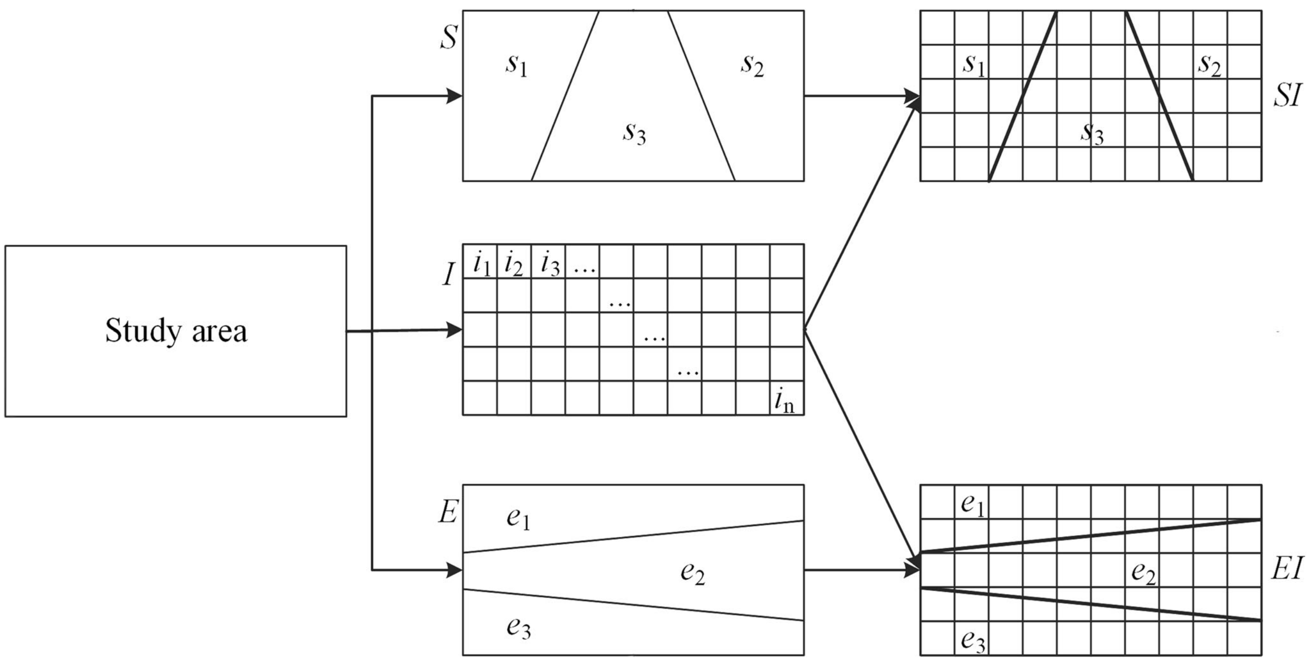

Fig. 2 The flowchart of GeogDetector

Table 1 Descriptive statistics of soil heavy metal concentration in soil (mg/kg)

	Min	Max	Median	Mean	CV	Background[a]	Standard[b]	Skewness	Kurtosis
As	2.63	43.20	7.07	8.99	0.69	6.88	30/25/20	2.309	6.222
Cd	0.05	1.42	0.20	0.27	0.81	0.17	0.3/0.3/0.6	2.431	6.846
Cr	10.50	104.00	54.40	52.90	0.38	55.99	150/200/250	− 0.024	− 0.572
Hg	0.03	0.50	0.11	0.13	0.62	0.17	0.3/0.5/1.0	1.602	3.073
Pb	17.00	62.60	30.90	31.66	0.38	35.70	300/300/300	0.720	0.454

CV coefficient of variation

[a]Data from Xu et al. (2012)

[b]Based on guidelines in the Environmental Quality Standard for Soils in China (GB 15618-1995) and the permissible limits for soils with pH less than 6.5, 6.5–7.5, and more than 7.5

This may be due to the combined effect of natural processes and a long history of industrial activities and urbanization (Fei et al. 2018a, Zang et al. 2017). The coefficient of variation (CV) of the heavy metals decreased in the order of Cd (81%) > As (69%) > Hg (62%) > Pb (38%) = Cr (38%). Additionally, according to the skewness and kurtosis values, and the Kolmogorov–Smirnov tests of normality ($P < 0.01$ for all heavy metals), the heavy metals had a non-normal distribution. High CVs and skewed distributions caused by a high degree of geochemical variation (differed greatly with respect to different sites), indicate that the heavy metal concentrations were strongly influenced by human activities (Shao et al. 2016; Mamut et al. 2018).

3.2 Pollution assessment

The *PI* values of each heavy metal in the agricultural soil samples are summarized in Table 2. The *PI* values of As, Cd, Cr, Hg, and Pb ranged from 0.38 to 6.28, 0.32 to 8.35, 0.19 to 1.86, 0.20 to 2.94, and 0.48 to 1.75 mg/kg, respectively. The mean *PI* values for each heavy metal in decreasing order were Cd (1.59) > As (1.31) > Cr (0.94) > Pb (0.89) > Hg (0.78). The *PI* values indicated that the study area was either unpolluted or slightly polluted by Pb and Cr ($PI \leq 2$). Although Hg had the lowest *PI* value, 2.88% of the soil samples exhibited mild pollution ($2 < PI \leq 3$). The level of As and Cd pollution was considerable; the percentages of mild pollution, moderate pollution ($3 < PI \leq 5$), and high pollution ($PI > 5$) were 8.05%, 4.88%, and 1.12%, respectively, for As, and 10.09%, 6.79%, and 3.40%, respectively, for Cd.

Table 2 The pollution index value of each heavy metal in soil

	Mean	Min	Max	Percentage of unpolluted and slightly polluted (%)
As	1.31	0.38	6.28	85.95
Cd	1.59	0.32	8.35	79.72
Cr	0.94	0.19	1.86	100
Hg	0.78	0.20	2.94	97.12
Pb	0.89	0.48	1.75	100

As for the *NIPI*, the values ranged from 0.59 to 6.39. Overall, 81.22% of the soils had a value indicating no pollution, pollution near the warning threshold, or low levels of pollution; 11.06% were moderately polluted and 7.72% were severely polluted. The results of the *PI* and *NIPI* indicated that almost one-fifth of the soil in the study area exhibited moderate pollution, with As and Cd being the main pollutants.

The *ER* values of each heavy metal in the agricultural soil samples are summarized in Table 3. The *ER* values of As, Cd, Cr, Hg, and Pb ranged from 3.82 to 62.79, 9.53 to 250.59, 0.38 to 3.71, 8 to 117.65, and 2.38 to 8.78, respectively. The mean *ER* values of each metal in decreasing order were Cd (47.52) > Hg (31.21) > As (13.07) > Pb (4.43) > Cr (1.89). According to the *ER* values of Cr and Pb, all the soil samples were in the low risk category. Although only 2.88% of the soil samples were classified as mildly polluted based on the *PI* value of Hg, 19.63% and 2.88% of soils were classified as moderate and considerable ecological risks, respectively, due to the high toxic-response factor of Hg (40). In the case of Cd, 58.14% of the soil samples were in the low risk category, while 28.79%, 10.14%, and 2.93% of the soils were in the moderate, considerable, and high risk categories respectively. In the case of As, despite its relatively high *PI* values, most of the soil samples (96.98%) were in the low risk category and 3.02% of the soils were in the moderate risk category because of its relatively low toxic-response factor (10).

The *RI* value ranged from 31.48 to 374.20. Overall, 88.24%, 11.47%, and 0.29% of the soil samples were in the low risk, moderate risk, and considerable risk categories, respectively. Cd and Hg had the highest contribution to *RI* (48.43% and 31.81%, respectively). The contributions of As, Pb, and Cr were 13.32%, 4.51%, and 1.93%, respectively. *ER* and *RI* are effective indicators of the degree of the individual and comprehensive ecological risk of soil heavy metals (Jiang et al. 2017b). The results showed that there is a moderate ecological risk of soil heavy metal pollution in the study area. Approximately 12% of the soil exhibited a moderate or higher ecological risk, with Cd presenting the highest ecological risk due to its high concentration and toxic-response factor (30). Moreover, Cd is easily absorbed by crops and can harm human health due to when ingested (Hu et al. 2017). Thus, Cd pollution of the soil present in the study area requires further attention.

Table 3 The ecological index value of each heavy metal in soil

	Mean	Min	Max	Percentage of low risk (%)
As	13.07	3.82	62.79	96.98
Cd	47.52	9.53	250.59	58.14
Cr	1.89	0.38	3.71	100
Hg	31.21	8	117.65	77.49
Pb	4.43	2.38	8.78	100

3.3 Identification of sources of heavy metals

According to the Spearman correlation analysis, the heavy metals were closely related to each other. The high homology between the heavy metals in agricultural soils indicated that they may have common sources such as lithogenic components, soil parent materials and agricultural activities (Gao et al. 2017). The contributions (q values) of the detected contribution factors (sources) on the distribution of heavy metals are shown in Table 4. The GeogDetector model also revealed that the soil parent material had a significant impact on the distribution of all heavy metals. The q values increased in the order of Hg (0.113) < Pb (0.125) < Cd (0.127) < As (0.164) < Cr (0.214). Many studies have reported that Cr originates from

Table 4 The q values of the influence factors on heavy metals in soil

	SPMs	FT	IP	NC	FU	PU
As	0.164**	0.112**	0.118**	0.135**	0.124**	0.122**
Cd	0.127**	0.002	0.004	0.121**	0.006	0.110**
Cr	0.214**	0.126**	0.125**	0.004	0.011	0.032
Hg	0.113**	0.015	0.110**	0.110**	0.032	0.032
Pb	0.125**	0.006	0.017	0.007	0.114**	0.110**

SPMs soil parent materials, *FT* farmland type, *IP* industrial production, *NC* number of cars (1/1000), *FU* fertilizer use, *PU* pesticide use

**Statistically significant at 0.01 level the soil parent material (Chen et al. 2016; Salonen and Korkka-Niemi 2007; Xue et al. 2014) and thus, the soil parent material showed the highest q value for Cr concentrations. Table 5 shows the concentrations of heavy metals in different soil parent materials. Generally, As, Cd, Hg, and Pb had the lowest concentrations in estuarine facies, whereas Cr had the lowest concentration in alluvial facies; As and Cd had the highest concentrations in eluvial facies, whereas Cr, Hg, and Pb had the highest concentrations in lacustrine facies.

Farmland types had a significant influence on the concentrations of As ($q = 0.112$) and Cr ($q = 0.126$). Farmland types with heavy metal contamination were, in increasing order of concentrations, paddy fields (9.58 and 51.98), dry land (10.24 and 56.19), orchards (13.91 and 58.53), and tea gardens (20.75 and 63.57) of mg/kg As and Cr, respectively. Statistical analyses showed that there were significant differences ($P < 0.05$) in As and Cr concentrations in paddy fields compared to tea gardens and orchards. This indicates that different types of agriculture have different influences on the concentrations of heavy metals in soil. Thus, by adjusting the type of agriculture, the pollution levels of As and Cr can be reduced, which can improve food safety and human health (Hu et al. 2017).

Industrial production had a significant influence on the concentrations of As ($q = 0.118$), Cr ($q = 0.125$), and Hg ($q = 0.110$). As a result of increased industrial production, the concentrations of As, Cr, and Hg increased from 4.81 to 12.53, 27.24 to 58.72, and 0.14 to 0.21 mg/kg, respectively (Fig. 3a). Many industrial activities, such as non-metallic mineral smelting, metallurgy, and chemical manufacturing, are carried out in the study area (Fei et al. 2018a). Previous studies reported that these industries cause the enrichment of As, Cr, and Hg in soil, through atmospheric deposition (Liang et al. 2017; Liu et al. 2017; Luo et al. 2015). Industrial activities had a huge impact on heavy metals in soil, particularly on As and Hg which demonstrated heavy pollution and high ecological risk according to the results discussed in the previous section. Therefore, industrial activities must be properly regulated and strictly limited to protect this area.

The number of cars per thousand people had a significant influence on the concentrations of As ($q = 0.135$), Cd ($q = 0.121$), and Hg ($q = 0.110$). With the increase in the number of cars per thousand people, the concentrations of As, Cd, and Hg increased from 6.92 to 13.87, 0.17 to 0.46, and 0.14 to 0.20 mg/kg, respectively (Fig. 3b). Heavy traffic causes air pollution, indicating that As, Cd, and Hg concentrations in soil are significantly influenced by atmospheric deposition, which agrees with the findings from previous studies (Engle et al. 2005; Liang et al. 2017; Micó et al. 2006).

Fertilizer use significantly influenced the concentrations of As ($q = 0.124$) and Pb ($q = 0.114$). With the increase in the use of fertilizers, the concentrations of As and Pb increased from 4.81 to 12.94, and 26.65 to 37.30 mg/kg, respectively (Fig. 3c). Pesticide use significantly influenced the concentrations of As ($q = 0.122$), Cd ($q = 0.110$), and Pb ($q = 0.110$). With the increase in the use of pesticides, the concentrations of As, Cd, and Pb increased from 4.81 to 14.15, 0.18 to 0.45, and 26.65 to 38.08 mg/kg, respectively (Fig. 3d). Although fertilizers and pesticides are essential for successful harvests, long-term use of the same fertilizers and pesticides can accumulate heavy metals in soils (Marrugo-Negrete et al. 2017). Previous studies confirmed that the long-standing farming practices of fertilizer and pesticide application can lead to the accumulation of heavy metals such as As and Pb in soils (Jiang et al. 2017b; Liu et al. 2017; Qiao et al. 2011). This is especially true for Cd, which is closely related to the intensive use of chemical fertilizers and pesticides, and is usually seen as a marker of agricultural activities (Wang et al. 2015).

Despite this, pesticides did not have a significant influence on all heavy metals. When considering the impact of the interaction of two source factors, the joint impact of the soil parent materials and pesticides exhibited the highest q values for all the heavy metals. The interaction q values were 0.316, 0.255, 0.279, 0.186, and 0.275 for As, Cd, Cr, Hg, and Pb, respectively (Table 6). Besides, the interaction q values were higher than the sum q values of soil parent materials and pesticide use which indicating that these two sources had a nonlinear enhancement. The reasons behind this phenomenon are very complicated that needed to be analyzed in further studies, for example, agricultural activities can accelerate the release of heavy metals in soil parent materials, diverse heavy metal contamination level is resulted by different types and strengths of agricultural

Table 5 The mean concentration (ppm) of heavy metals in various soil parent materials

	Lacustrine facies	Estuarine facies	Alluvial facies	Eluvial facies
As	7.52	5.51	8.96	14.19
Cd	0.19	0.17	0.39	0.41
Cr	72.87	58.13	46.83	53.55
Hg	0.21	0.14	0.15	0.14
Pb	35.23	27.27	35.1	34.82

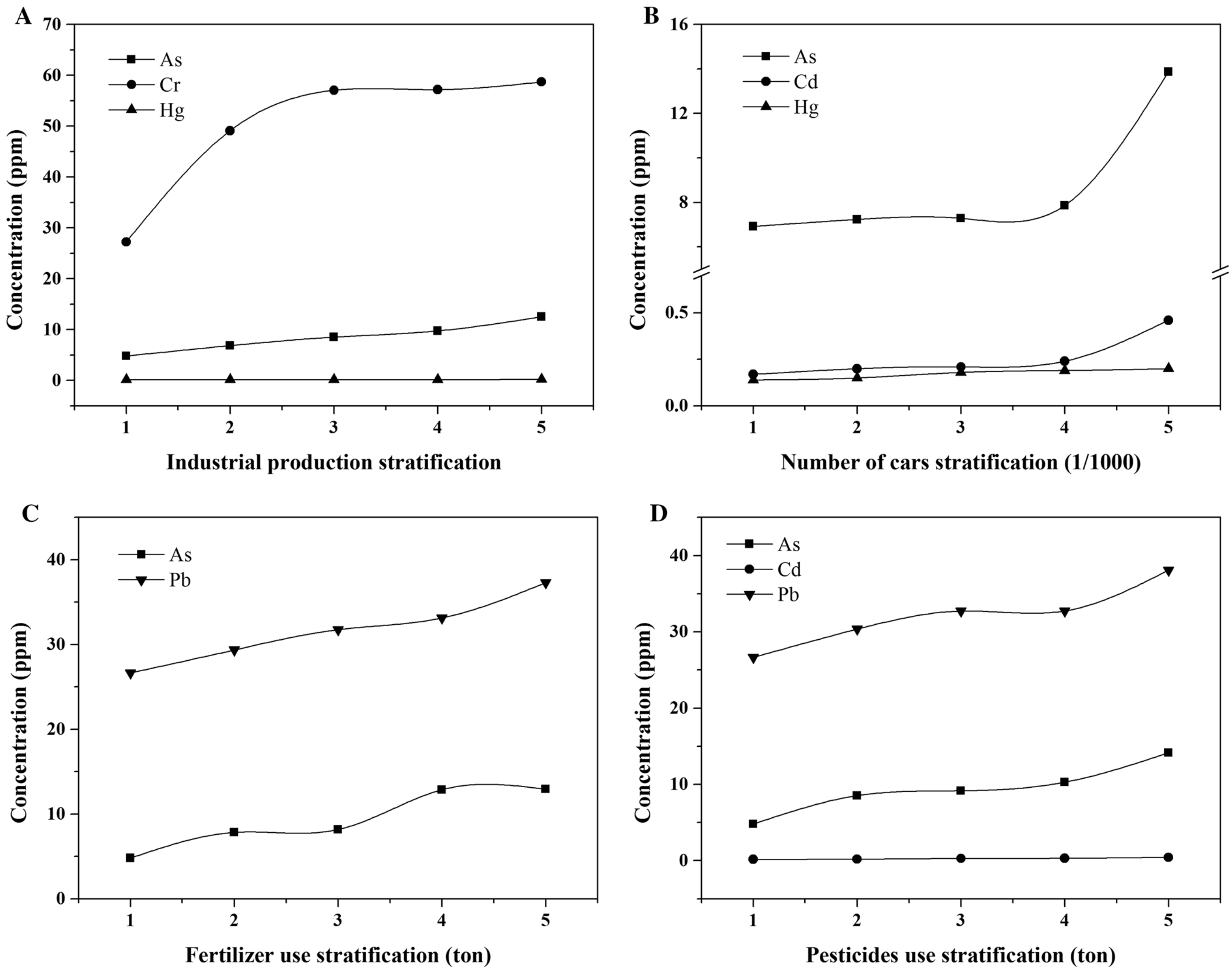

Fig. 3 The concentrations of heavy metals from stratification of different source factors (1–5 corresponded to low, mild, moderate, high, and very high stratification respectively)

Table 6 The max interaction q values on various heavy metals in soil

	Interaction factors	q value
As	SPMs & PU	0.316
Cd	SPMs & PU	0.255
Cr	SPMs & PU	0.279
Hg	SPMs & PU	0.186
Pb	SPMs & PU	0.275

SPMs soil parent materials, *PU* pesticides use, non-linear enhancement: interaction q values greater than the sum of their respective q value

activities on various soil parent materials, etc. These interaction results indicated that natural processes and agricultural activities were the main sources of soil heavy metal pollution in the study area.

4 Conclusions

This study quantitatively assessed the pollution level, ecological risk, and sources of heavy metals in agricultural soils in a typical coastal industrial area, undergoing rapid industrialization and urbanization. According to the results, Cd was the most polluting element in the soil, and, due to its high toxic-response factor, it also poses the highest risk for ecological systems and human health. Another element responsible for heavy pollution was As. However, due to its relatively low toxic-response factor, most of the soils were in the low ecological risk category. In contrast, although Hg contamination had relatively small concentrations, one-fifth of the soil had a moderate ecological risk due to its high toxic-response factor. A novel geographical detector model was employed to quantitatively identify the different sources of heavy metals. Natural processes (soil parent material) had a significant impact on all the heavy metals analyzed in this study. Additionally, farmland types had a significant influence on Cr and As, industrial activities had a significant influence on As, Cr, and Hg, traffic significantly influenced As, Cd, and Hg concentrations, and agricultural applications of fertilizer and pesticide had a significant impact on As, Cd, and Pb. Arsenic, with diverse sources, requires further analysis in future studies. Natural processes and agricultural activities were found to be the main sources of soil heavy metal pollution in the study area.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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