Using multi-medium factors analysis to assess heavy metal health risks along the Yangtze River in Nanjing, Southeast China

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Abstract
In the environmental ecosystem, there are no absolutely isolated risks. Each risk might be influenced by multiple environmental factors and the factors’ interaction within the specific system. Hence, health risk assessments of heavy metal contamination must consider multiple environmental media and their transfer processes from one medium to another. Integrated assessments provide a new perspective for evaluating many factors, such as the potential ecological risks of soils, sediments, plants, and the transportation of heavy metals in these media, which influences the health risks. In this study, the main influencing factors for human health risk from heavy metals along the Yangtze River in Nanjing, Southeast China, were explored. The contents of five heavy metals were measured in sediment-soil-plant, including cadmium (Cd), lead (Pb), copper (Cu), zinc (Zn), and chromium (Cr). The Cd displayed the highest potential ecological risk in soils and sediments, as it possessed high bioaccessibility (BA; 0.17 ± 0.211) and bioaccumulation factor (BCF; 0.35 ± 0.33). The 5.97% of the target hazard quotient (THQ) of Cd were higher than 1, indicating a potential health risk in plant consumption. Based on the geodetector model, determinant power (DP) valves for factors influencing health risk strongly suggest that plant types (0.479) has a highest effect, followed by soil organic matter (SOM; 0.292), and the BA of heavy metals (0.107). The results also indicate that pollution from the upper reaches of the river, and agricultural activities, had a greater impact on health risk than did industrial activities in the study area. Thus, regular monitoring and source control for Cd along with integrated agricultural management practices should be implemented to control and reduce heavy metal inputs and improve the safety of cultivated plants.

1. Introduction

Heavy metal contamination has been accelerating worldwide over the last two decades owing to rapid economic development and industrialization (Facchinelli et al., 2001; Solgi et al., 2012; Zang et al., 2017). Prolonged excessive intake of heavy metals may lead to their chronic accumulation in the kidney and liver of humans, and has been linked to nervous, cardiovascular, kidney, and bone diseases (Li et al., 2015). Previous studies have also found that a high incidence of stomach cancer is closely related to high consumption of cadmium (Cd), lead (Pb), copper (Cu), chromium (Cr), and other metals found in soil, fruit, and vegetables (Türkdoğan et al., 2003). Therefore, it is necessary to assess health risks associated with heavy metals and identify the influencing factors in order to improve protection of the environmental and of human health.

Efforts have been made to properly investigate the potential risks to human health associated with heavy metals, and a large body of research has reported levels of heavy metal contamination in multiple environmental media, including soils, sediments, water, and plants (Schreck et al., 2013). These studies assessed heavy metal risk and pollution sources based on the spatial distribution of the contaminants. The accumulation of heavy metals in multiple media have been found to be the result of natural and anthropogenic factors (Luo et al., 2011; Chi et al., 2017; Jiang et al., 2017; Xu et al., 2018). However, environmental ecology is focused on interrelated system, so the health risk is influenced by multiple
environmental media and their interrelated transfer processes. The fate and transport of heavy metals taken up by plants from the soil are two of the most important mechanisms that affect human intake of these toxic contaminants (Hu et al., 2014; Liu et al., 2017; Zhang et al., 2018).

As one of the most economically developed cities in China, Nanjing is located on the middle and lower part of the Yangtze River and has a complex industrial structure (Wang et al., 2016), rapid development of urbanization, and a heavy utilization of soil agriculture. The Yangtze River is the longest river in China and the third longest river in the world, it plays a crucial role in China's sustainable economic and social development (Li et al., 2012). The Yangtze River is also a pathway for contaminant migration and exposure (Shao et al., 2016). Heavy metals that accumulated in the local atmosphere had already been shown to cause potential health risks (Li et al., 2015). The quality of the study area has attracted widespread public attention, leading to the need for more extensive assessment of risks and more frequent contaminant monitoring to ensure the health of the surrounding environment and the residents.

The primary objective of this study was to develop an integrated risk assessment methodology to assess heavy metal pollution by identifying and characterizing a detailed list of environmental media and factors that affect health risk in the Yangtze River basin using a geodetector model. Geodetector models were developed by Wang et al. (2010), and that have displayed advantages in the diagnosis of issues that affect the spatial distribution of contaminants (Hu et al., 2014). Since very few assumptions and constraints are used in this method, the limitations of traditional statistical analysis methods in dealing with geospatial problems associated with categorical variables can be effectively overcome (Wang and Xu, 2017). The aims of this study were: 1) to measure the accumulated concentrations of heavy metals in different types of soils, sediments, and plants in study area; 2) to assess heavy metals with a potential ecological risk index for each environmental medium and a risk exposure model recommended by the United States Environmental Protection Agency (USEPA) to characterize risk to human health; 3) to analyze the heavy metal transfer mechanisms in sediment-soil-plant systems, based on soil profile distributions, the bio-accessibility (BA) of heavy metals, and estimated bioaccumulation factors (BCFs); and 4) to quantify the factors that affect health risk, based on multiple media in the environment.

2. Materials and methods

2.1. Study area and sampling

The study was focused on lower reaches of the Yangtze River (31°14′ – 37°32′ N, 118°22′ – 119°14′ E), mainly in Nanjing, Jiangsu, China (Fig. 1). The middle and lower reaches of the Yangtze River are characterized by Paleozoic marine source rocks and Quaternary unconsolidated sediment. The main mineral resources are Cu (Wang et al., 2009), Zn and Pb (Zhang et al., 2012). The main land use type in the upper reaches of the study region is farmland; the middle reaches are primarily used for building and industry, and the lower reaches are industrial land and farmland. The main plants in the study area include rice, wheat, cotton, and vegetables.

A variety of sample types were collected as part of this study, including 124 surface soil samples (0–20 cm), 18 profile soil samples (20–40 cm, 40–60 cm, 60–80 cm, 80–100 cm), 75 sediment samples, and 67 typical plant samples that were collected with a wooden spatula during a low flow period in 2012. All sampling sites were geo-located using a global positioning system receiver. Five samples were collected within a area of 20 m² around each sampling site and mixed well to obtain a composite sample. All samples were bagged, labeled, and returned to the laboratory in clean polyethylene plastic bags placed in a cooler with a temperature of 4 °C (Lasorsa and Casas, 1996). Of all collected soil samples, 57 were from uncultivated lands, 30 were from wheat fields, 13 were from paddy fields, and 24 were from vegetable fields. A total of 75 sediment samples were collected along streams (15 samples) and Yangtze River (60 samples). All soil and sediment samples were air-dried, crushed, and then sieved through a 0.149 mm plastic sieve. Plant samples included 30 wheat samples, 13 rice samples, and 24 vegetable samples. Of all vegetable samples, seven were Chinese cabbage (Brassica chinensis var. Chinensis), four were garden radish (Raphanus sativus L. var. sativus), eight were Artemisia selengensis (Artemisia selengensis Turcz ex Bess. var. selengensis), and the remaining five were pepper (Capsicum annuum var. grossum). Plant samples were thoroughly washed with tap water and then rinsed with deionized water. After drying at 25 °C, vegetable samples were weighted and then further dried at 85 °C for 30 min; after drying, samples were kept in an oven at 60 °C until they reached a constant weight, at which point their dry weight was recorded. The content of plant moisture was determined from the difference in fresh and dry weights (Zhang et al., 2018). The dried samples were ground up and then stored in sealed plastic bags at room temperature for heavy metal analysis.

2.2. Chemical analysis

The soil and sediment properties of pH were measured using a soil/water ratio of 1:2.5. The pH was tested with a standard pH meter (PHS-3C, Shanghai, China; Huang et al., 2006), and soil and sediment organic matter (OM) was determined using the Walkley-Black method (Nelson et al., 1996).

The total content of heavy metals for soil and sediment samples, including Cd, Pb, Cu, Zn, and Cr were digested following the HNO3–HClO4 – HF method (Hu et al., 2018). The soil availability contents of heavy metals in wheat, paddy, and vegetable fields were evaluated by extracting metals in the soils with CaCl2 in accordance with Morgan and Alloway (1984). Using this method, a useful index of biological availability for these metals could be obtained (Andrews et al., 1996; Mcbride et al., 2004; Liang et al., 2017). A 20 ml sample of 0.05 M CaCl2 solution was added to a 50 ml centrifuge tube containing a 10 g soil sample. The suspension was thensubjected to linear shaking at room temperature for 30 min. Finally, the sample was centrifuged at 3200 rpm for 5 min to separate the supernatant. A total of 0.1 g of plant sample was prepared for digestion in mixed with ultrapure acid (HNO3: H2O2 = 2:1, in volume). The digested samples were diluted to 50 ml with ultrapure water (Kazi et al., 2006).

The contents of heavy metals were determined by inductively coupled plasma mass spectroscopy (ICP-MS; American Thermo Scientific, X7). The detection limits (in mg kg⁻¹) for the analyzed metals were: Cd, 0.03; Pb, 2.0; Cu, 0.5; Zn, 0.5; and Cr, 2.0 (Hu et al., 2016). Duplicate samples, reagent blanks, and standard reference materials (GBW07363 and GBW07429; the Center of National Standard Reference Material of China) were applied to guarantee analytical precision, with samples collected in triplicate to ensure the accuracy of the experiment. Lettuce was used as the plants' reference material (IPE 776, Wepal). The recovery of standard reference materials ranged from 94% and 103% for all test parameters. The concentrations of heavy metals detected in sediment and soil samples were consistent with the reference values, and the relative standard deviations (RSDs) for the replicate samples were less than 5%.
2.3. Data analysis

2.3.1. Traditional statistical analysis
Statistical analyses were carried out using the SPSS 21.0 (IBM Corporation, NY, USA). Standard deviation, coefficient of variation (CV), maximum, and minimum were calculated for Cd, Pb, Cu, Zn, Cr, OM, and pH. Analysis of variance (ANOVA) was performed by a Kruskal–Wallis test to identify significant differences in the potential ecological risks of heavy metals in soils and sediments. Furthermore, the health risks associated with the consumption of different plants containing heavy metal contamination, and the transfer processes of heavy metals in the soil-plant systems were also assessed via a Kruskal–Wallis test with a P level of 0.05.

2.3.2. Potential ecological risk index of heavy metals
The potential ecological risk index proposed by Hakanson (1980) and based on the concentrations, types, toxicity, sensitivity and background values of a heavy metal, was employed to assess ecological risks in a variety of research domains (Xie et al., 2013). In this study, this risk index was applied to assess the ecological risks of heavy metals in soil and sediment. The formula is expressed as follow:

\[
C_I^f = \frac{C_I}{C_n} \tag{1}
\]

\[
E_I^f = T_{EF}^f \times C_I^f \tag{2}
\]

\[
RI = \sum E_I^f = \sum T_{EF}^f \times C_I^f \tag{3}
\]

where \(C_I^f\) is the individual heavy metal contamination index; \(C_I\) is the concentration of each heavy metal in surface soil and surface sediment; \(C_n\) is the reference value for heavy metals, which was defined by the background value in Nanjing (Nanjing BGV); \(E_I^f\) is the monomial potential ecological risk factor; \(T_{EF}^f\) is the heavy metal toxic response factor, which is 30, 5, 5, 1, and 2 for Cd, Pb, Cu, Zn and Cr, respectively (Hakanson, 1980); and \(RI\) is the potential ecological risk of the overall contamination.

The five categories of \(E_I^f\) were: low risk (less than 5), moderate risk (between 5 and 10), considerable risk (between 10 and 20), high risk (between 20 and 40), and very high risk (greater than 40). The four classes of \(RI\) were identified as: low risk (less than 30), moderate risk (between 30 and 60), considerable risk (between 60 and 120), and very high risk (greater than 120; Yuan et al., 2015).

2.3.3. Health risk assessment of heavy metals
The target hazard quotient (THQ) is an index established by using version III risk-based concentration tables established by the US Environmental Protection Agency (USEPA, 2015) to assess the health risks to populations; it can simultaneously assess the health risks caused by the presence of a single heavy metal or multiple heavy metals (Chen et al., 2013; Hu et al., 2017; Tepanosyan et al., 2017). This method assumes that the human body’s absorption dose of the pollutant is the same as the ingested dose, with the ratio of ingested dose and reference dose defined as the evaluation criteria. If the ratio is less than 1 (THQ < 1), there is no obvious health risk for people exposed to pollutant; otherwise, a health risk exists. This relationship is expressed as:

\[
THQ = \frac{EF \times ED \times IR \times C_m \times 10^{-3}}{B_w \times AT \times RfD} \tag{4}
\]

where \(EF\) is the exposure frequency (365 d year\(^{-1}\)); \(ED\) is the daily plant ingestion, which is considered to be 238.3 g person\(^{-1}\) d\(^{-1}\) of rice, 140.2 g person\(^{-1}\) d\(^{-1}\) of wheat, and 276.2 g person\(^{-1}\) d\(^{-1}\) of vegetables (CNEPA, 2013); \(C_m\) is the heavy metal concentration in plants (mg kg\(^{-1}\), on fresh weight basis); \(B_w\) is an average body weight of an adult (60.6 kg person\(^{-1}\); CNEPA, 2013); \(AT\) is the average exposure time (365 d year\(^{-1}\) × number of exposure years, assumed to be 70 years for this study; Hu et al., 2017); and \(RfD\) is the reference dose (mg kg\(^{-1}\) d\(^{-1}\)), which is regarded as an estimate of daily exposure to human population (USEPA, 2015). The values of \(RfD\) for Cd and Zn were 0.001 and 0.3 mg kg\(^{-1}\) d\(^{-1}\), respectively, as obtained from the US EPA Integrated Risk Information System (USEPA, 2015). The values of \(RfD\) for Pb and Cu were 0.004 and 0.04 mg kg\(^{-1}\) d\(^{-1}\), respectively, as obtained from China’s National Environmental Protection Agency (CNEPA, 2009).

The hazard index (HI) is expressed as the sum of THQs associated with each exposure route, expressed as:

\[
HI = \sum_{i=1}^{n} THQ \tag{5}
\]

The chronic toxic effect is defined as \(HI > 10\).

2.3.4. Transfer of heavy metals from different environmental media
Bioaccessibility of heavy metals (BA) can be expressed as the
ratio of soil-available heavy metals to total heavy metals; it is considered a better indicator of the impact of heavy metal contamination in soil (Liu et al., 2017). These equations can be expressed as:

\[ BA = \frac{C_{\text{available}}}{C_{\text{total}}} \]  

(6)

\[ TBA = \sum_{i=1}^{n} BA_i \]  

(7)

where \( C_{\text{available}} \) and \( C_{\text{total}} \) are available heavy metal concentrations (mg kg\(^{-1}\)) and total metal concentrations in soil (mg kg\(^{-1}\)), respectively; and \( TBA \) is the sum of all \( BA \)s for the heavy metal.

The bioaccumulation factors (BCF) for different types of plants were calculated to identify the potential capability of transmission of heavy metals from soil to the edible parts of plants (Yang et al., 2017). These equations can be calculated to evaluate the factor’s spatial distribution expressing as:

\[ BCF = \frac{M_p}{M_s} \]  

(8)

where \( M_i \) is total heavy metal concentrations in soil (mg kg\(^{-1}\)), and \( M_p \) is the total metal concentrations in fresh plants (mg kg\(^{-1}\)).

### 2.3.5. Factors affecting the health risks of heavy metals

The distribution of most geographical characteristics and their influencing indicators on a spatial scale generally obey a certain rule; specifically, if there is a similar spatial distribution pattern between a geographical characteristic and a factor, this indicates that there is a direct or indirect relationship between the factor and the geographic characteristic, and the determination power \( (DP) \) can be calculated to evaluate the factor’s spatial distribution effect with respect to the geographic characteristic. To analyze the spatial relationship between \( Y \) (geographic characteristic) and \( X \) (factors), the strata of \( Y \) and \( X \) are overlaid, as depicted in Fig. S1 (Wang et al., 2010). The mean values and variances of \( Y \) for strata of \( X \) are represented by \( \bar{Y}_{i1}, \bar{Y}_{i2}, \bar{Y}_{i3} \) and \( \sigma_{i1}^2, \sigma_{i2}^2, \sigma_{i3}^2 \), respectively.

Using a statistical method to test the significant differences between \( \bar{Y}_{i1}, \bar{Y}_{i2}, \bar{Y}_{i3} \); the \( DP \) of \( X \) to \( Y \) can be expressed as:

\[ P_{D,U} = 1 - \frac{1}{n_0 U} \sum_{i=1}^{m} n_{D_i} \sigma_{U_{i}}^2 \]  

(9)

where \( P_{D,U} \) is the \( DP \) of \( X \) to \( Y \), \( n_{D_i} \) is the number of samples in strata of \( X \), \( m \) is the number of samples in whole study region, \( n_0 \) is the number of grade regions, \( \sigma_{U_{i}}^2 \) is the variance of \( Y \) in the entire zone, and \( \sigma_{D_i}^2 \) is the variance of \( Y \) in strata of \( X \). Assuming \( \sigma_{D_i}^2 \neq 0 \), the model was constructed. \( P_{D,U} = 1 \) indicates \( Y \) was completely affected by the partition factor; \( P_{D,U} = 0 \) means the distribution of \( Y \) was random; generally, the value of \( P_{D,U} \) was between 0 and 1.

Larger the values indicate a greater influence of \( X \) on \( Y \). In the present study, the factor detector of this model was used to detect \( DP \) for health risk based on the transfers and risks from environmental media. So, the health risk was \( Y \), and the above factors were \( X \). The data were preprocessed in ArcGIS 10.0. According to the input requirements of the geodetector model, the projection was unified with the GCS_WGS_1984 projection coordinate system. Vector datum elements were obtained by interpolating the analyzed values of the sample locations, then divided through a geometrical interval method (Cao et al., 2013), and finally exported to raster datum.

### 3. Results and discussion

#### 3.1. Accumulation of heavy metals in different environmental media

The accumulations of heavy metals in soils, sediments, and plants are presented in Table 1. The mean value of pH was 7.12 in soils and 7.70 in sediments. The mean content of OM in soils (21.32 g/kg) was higher than in sediments (16.44 g/kg). The mean sediment concentrations of the heavy metals Cd, Pb, Cu, Zn, and Cr were 0.66, 41.90, 46.58, 122.86, and 84.93 mg/kg, respectively, all of which were higher than those detected in soil. Previous research has established that heavy metals have a strong affinity for sediments, which greatly impacts their mobility (Fang et al., 2016). The mean available soil concentrations of the heavy metals Cd, Pb, Cu, Zn, and Cr were 0.07, 0.01, 0.19, 0.87, and 0.05 mg/kg, respectively. The mean plant contents of the heavy metal Cd, Pb, Cu, Zn, and Cr were 0.08, 0.18, 4.19, 21.54, and 1.45 mg/kg, respectively. The CV values for soil available content of Cd, Pb, and Zn ranged from 112% to 146%, all indicating strong variation; similarly, the CV values for plant contents of Pb (116%) and Cr (115%) also indicated strong variations (Nielsen and Bouma, 1985). Detailed statistical descriptions of heavy metal contamination for soil, sediments and plant samples are listed in Table S1.

#### 3.2. Environmental risks of heavy metals in different environmental media

##### 3.2.1. Potential ecological risks of heavy metals in soils and sediments

The results of the potential ecology risk assessment for soils and sediments are presented in Fig. 2, based on the Nanjing BGV. The mean soil ecology risk values with standard derivations for Cd, Pb, Cu, Zn, and Cr were 44.17 ± 26.70, 5.89 ± 2.96, 6.17 ± 1.79, 1.37 ± 0.44, and 2.15 ± 0.53, respectively; the mean sediment ecology risk values with standard derivations for Cd, Pb, Cu, Zn, and Cr were 98.70 ± 47.75, 66.99 ± 2.95, 7.35 ± 1.99, 1.52 ± 0.37, and 2.21 ± 0.41, respectively. The mean RI values with standard derivations for soils and sediments were 59.75 ± 29.31 and 116.47 ± 51.05, respectively. According to standard classifications, Cd presented a very high degree of risk (\( E'_i \), value over 40), Pb and Cu presented moderate risk (\( E'_i \) values between 5 and 10), and the remaining heavy metals investigated in this study presented a low risk (\( E'_i \), value less than 5).

The Kruskal–Wallis test was used to identify significant differences at the 0.05 significance level (\( P < 0.05 \)). The \( E'_i \) value for the Cd of sediment samples along the Yangtze River was significantly higher than for other sites. The \( E'_i \) value for the Pb of sediments along the Yangtze River was significantly higher than wheat field soils. Likewise, the \( E'_i \) values for Cu and Zn in sediments along Yangtze River were also significantly higher than those wheat field soils and uncultivated land. Additionally, the \( E'_i \) value for the Cr in vegetable soils was significantly higher than wheat soils. The RI values of sediments along the Yangtze River were significantly higher than other sites. The accumulation of heavy metals in sediment is primarily caused by heavy metal pollution along the upper reaches of the Yangtze River; this pollution migrates to the study area with the river flow (Yi et al., 2011). Previous studies have found that substantial amounts of Pb in the environment are associated with higher abundances of Pb in parent rocks, making it more likely to be transferred to sediments and soils through the pedogenic process of the Yangtze River (Wang et al., 2013) and from atmosphere emissions (Feng et al., 2011; Huang et al., 2015).
The spatial distribution of soil RIs for the study area indicated that the high RI values were distributed in the Bagua and Jiangxin isles, as well as on the upper reaches and lower reaches of the Yangtze River (Fig. 3a). Bagua isle and Jiangxin isle have been used as farmland for decades. The high RI of each isle was caused by the high content of heavy metals Cu and Cd. Previous research has found that the distributions of Cu can be attributed to geochemical background phenomena on a regional scale in the Yangtze River delta region, and significant Cd and Pb contaminant concentrations have been found to be distributed throughout the upper reaches of the Yangtze River (Shao et al., 2016). The spatial distribution of sediment RIs in the study area is shown in Fig. 3b. Considerable or very high-risk sediment samples were found in the study area. The RI value for sediment samples along Yangtze River was relatively higher than that along the streams.

### 3.2.2. Health risks of heavy metals from consumption of different plants

The potential health risks associated with the consumption of different plants containing Cd, Pb, Cu, Zn, and Cr were assessed based on THQ and HI (Fig. 4). Mean values ± standard derivations of THQ for Cd, Pb, Cu, Zn, and Cr caused by human consumption of local plants were 0.28 ± 0.33, 0.16 ± 0.21, 0.33 ± 0.22, 0.20 ± 0.12, and 0.003 ± 0.004, respectively. The mean value of HI was estimated at 0.97 ± 0.67. These results indicate that potential health risks of Cu and Cd are relatively higher than those of the other

### Table 1

Descriptive statistical results for heavy metal contamination of different environmental media.

<table>
<thead>
<tr>
<th>Medium</th>
<th>Parameter</th>
<th>pH</th>
<th>OM</th>
<th>Cd</th>
<th>Pb</th>
<th>Cu</th>
<th>Zn</th>
<th>Cr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil (n = 124)</td>
<td>Minimum</td>
<td>4.58</td>
<td>2.78</td>
<td>0.10</td>
<td>12.70</td>
<td>14.30</td>
<td>51.60</td>
<td>16.70</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>8.91</td>
<td>48.05</td>
<td>1.20</td>
<td>150.00</td>
<td>86.60</td>
<td>356.00</td>
<td>232.00</td>
</tr>
<tr>
<td></td>
<td>Mean</td>
<td>7.12</td>
<td>21.32</td>
<td>0.29</td>
<td>36.87</td>
<td>39.13</td>
<td>110.80</td>
<td>82.65</td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
<td>0.94</td>
<td>9.26</td>
<td>0.18</td>
<td>18.51</td>
<td>11.35</td>
<td>35.57</td>
<td>20.54</td>
</tr>
<tr>
<td>Sediment (n = 75)</td>
<td>Minimum</td>
<td>5.58</td>
<td>1.98</td>
<td>0.16</td>
<td>15.40</td>
<td>16.00</td>
<td>56.10</td>
<td>49.30</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>8.41</td>
<td>53.76</td>
<td>1.47</td>
<td>132.00</td>
<td>78.90</td>
<td>183.00</td>
<td>140.00</td>
</tr>
<tr>
<td></td>
<td>Mean</td>
<td>7.70</td>
<td>16.44</td>
<td>0.66</td>
<td>41.90</td>
<td>46.58</td>
<td>122.86</td>
<td>84.93</td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
<td>0.34</td>
<td>8.77</td>
<td>0.32</td>
<td>18.44</td>
<td>12.59</td>
<td>29.51</td>
<td>15.62</td>
</tr>
<tr>
<td>Soil (available) (n = 67)</td>
<td>Minimum</td>
<td>0.04</td>
<td>0.53</td>
<td>0.48</td>
<td>0.44</td>
<td>0.27</td>
<td>0.24</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>Maximum</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
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<td>--</td>
<td>--</td>
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<tr>
<td></td>
<td>Mean</td>
<td>--</td>
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<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
<td>0.007</td>
<td>0.001</td>
<td>0.13</td>
<td>1.36</td>
<td>0.04</td>
<td>0.04</td>
<td>0.04</td>
</tr>
<tr>
<td>Plant (n = 67)</td>
<td>Minimum</td>
<td>0.43</td>
<td>0.82</td>
<td>8.87</td>
<td>57.00</td>
<td>7.30</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mean</td>
<td>0.08</td>
<td>0.18</td>
<td>4.19</td>
<td>21.54</td>
<td>1.45</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
<td>0.07</td>
<td>0.21</td>
<td>2.42</td>
<td>15.31</td>
<td>1.67</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>CV</td>
<td>0.90</td>
<td>1.16</td>
<td>0.58</td>
<td>0.71</td>
<td>1.15</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>CV</td>
<td>0.08</td>
<td>0.21</td>
<td>2.42</td>
<td>15.31</td>
<td>1.67</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nanjing BGV</td>
<td>CV</td>
<td>0.20</td>
<td>3.28</td>
<td>31.69</td>
<td>80.71</td>
<td>76.95</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup> CV—Coefficient of variation.

<sup>b</sup> The background values (BGV) of Nanjing were adapted from the study of [Wu, 2005].
heavy metals for residents in the study area. Mean values of THQs were below the safe threshold value of 1. Only 5.97% of the THQs of plant samples were higher than 1 for Cd representing a potential health risk with respect to the plants’ consumption. There is no specified maximum level of contaminants in food (MLCF) for Cu in current Chinese standards (SEPAC, 2017). The THQ value for the Cd in rice was significantly higher than for vegetable (P value < 0.05), and the THQ value for the Pb in rice was significantly higher than for vegetable (P value < 0.001) and wheat (P value < 0.001). The HI for heavy metals of rice was significantly higher than for vegetable (P value < 0.001) and wheat (P value < 0.001). The reason for the differences in HI and THQ is the different metal uptakes among rice, wheat, and vegetables (Khan et al., 2013). It has been reported that heavy metal accumulation in plants depends upon plant type and the efficiency of different plants in their absorption of heavy metals; this is evaluated either by plant uptake or soil-to-plant transfer factors of the heavy metals (Rattan et al., 2005; Hu et al., 2017).

The spatial distributions of health risk for different plant types in the study area are shown in Fig. 5. Wheat fields are most widely distributed in the suburbs of Nanjing, along the Yangtze River. The paddies are distributed on Bagua isle and the upper reaches of the study area. Vegetable fields are mainly distributed on Bagua isle. The HI values of wheat do not indicate an obvious spatial distribution trend. This suggests the potential ecological risks for soil and sediments associated with wheat could not be accurately inferred from the migration of heavy metals or the extent of the damage to human health. The potential ecological risk associated with vegetable soils may be caused by fertilization, but the HI indicated low sensitivity to the potential ecological risks associated with these soils. Fertilizer application may not have a direct effect on the accumulation of heavy metals in plants; thus, it may not accurately
reflect the spatial distribution of HI values in the study area. Different conditions were observed in paddy field soils and dry land soils, and different pH values in these areas may lead to different conversion efficiencies for heavy metals (Liu et al., 2016). Therefore, it is necessary to further study the conversion efficiency of the available heavy metals.

3.3. Transfer processes of heavy metals in the multi-medium systems

The BA and BCF of heavy metals for the soil-plant system were calculated. The orders of the BA and BCF were Cd > Zn > Cu > Cr > Pb. The mean values ± standard derivations of BA for Pb and Cd were 0.0003 ± 0.004 and 0.17 ± 0.211, respectively; the mean values ± standard derivations of BCF for Pb and Cd were 0.005 ± 0.006 and 0.35 ± 0.33, respectively. The mean value ± standard derivation of TBA was 0.19 ± 0.22. Spearman correlations for individual variables for heavy metal transfer are summarized in Table 2. The results indicate that SOM has a significant correlation with pH (P < 0.05) and soil RI (P < 0.01); HI has a significant correlation with BA (P < 0.05), and sediment RI (P < 0.01). It is necessary to evaluate the determination power of sediment RI to health risk. However, all sediment samples were along the Yangtze River. Interpolation of sediment samples for the whole study area would not be objective. Thus, distances from the river to HI of sampling points represent the determination power of sediment RI which, in turn, should be one of main HI factors. Soil pH and organic matter have been reported to influence the transfer of heavy metals in soil-plants systems. Soil pH not only influences the bioavailability of heavy metals in the soil (Willscher et al., 2017), but also further influences their availability and toxicity to plants (Bravo et al., 2015). Levels of organic matter in soils also affect plant uptake of heavy metals and/or migration in groundwater by immobilizing them in soil (Kwiatkowska-Malinia, 2018). Combined with the results of spearman correlation analysis, the SOM and pH might indirectly affect HI values.

Soils in the Yangtze River area are high maturity, they have been developed from the marine strata and are influenced by the Yangtze River alluvial deposits to a significant degree (Soil Survey Office of Jiangsu Province of China, 1995). Sedimentation processes typically occur during the subsequent transport of heavy metals from anthropogenic sources (Liu et al., 2001). The soil profile distribution of pH, SOM, and heavy metals is illustrated in Fig. 6. A gradual decrease was seen in soil pH from topsoil to the subsoil suggesting acidification in surface soil layers. The change trend of SOM content is opposite to the change trend of pH from the topsoil soil to the subsoil. The heavy metal pollution of industrial zones was higher than for other areas. The contents of Cd, Pb, Cu, Zn, and Cr were 0.56–0.64, 44.3–58.9, 53.3–69.2, 127–150, and 80.1–80.2 mg/kg, respectively. These results suggest that the heavy metal contamination of soils was influenced by anthropogenic factors such as agricultural and industrial activities in study area. However, heavy metal contamination was still noticeable in subsoil layers indicating that topsoil accumulation of heavy metals in this region was affected by the superimposition of industrial activities and the environmental background of the river basin. The influence of industrial activities on heavy metals’ accumulation was not obvious. To further verify the determination power of industrial activities to HI, the distance from industrial areas has been added as a factor.

3.4. Factors influencing the health risks of heavy metals

Based on our results, it can be concluded that the soil RI, BA, pH, SOM, plant types, distance from the river, and distance from industrial areas have different degrees of influence on the spatial distribution of HI. All of the factors were interpolated and divided into eight classes based on geometrical interval method (Wang et al., 2010; Cao et al., 2013). The geodetector model was employed to quantify the distribution relationship between HI and the above factors. The classification information of the raster datum was extracted to each plant sample location with HI values (Fig. 52). Plant types were divided into three categories, including vegetables, rice, and wheat.

Using the geographical detector model, the determinant power values of the above factors on influencing health risk were calculated. As shown in Fig. 7, the DP value of the factors displaying a marked difference can be ranked as follow: plant types (0.479) > SOM (0.292) > BA (0.107) > distance from the river (0.068) > soil RI (0.061) > pH (0.055) > distance from industrial areas (0.045). The results indicate that plant types, which displayed the highest DP value, can predominantly explain the spatial distribution of HI, followed by the SOM and BA of heavy metals. Thus, the results obtained using the geodetector model were more intuitive and accurate than traditional statistical analyses.

The determination power of distance from the river on HI was higher than that of soil RI and distance from industrial areas. This suggests that the pollution reaches from the upper part of the river and agricultural activities had a greater impact on the HI than do anthropogenic factors, especially industrial activities conducted in recent years. Despite the major influence of pH on metal speciation and/or metal toxicity to the organisms, it had a weak or indirect influence on health risk. This might be due to the health risk

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Table 2

Spearman correlation analysis of variables in the multi-medium system.

<table>
<thead>
<tr>
<th></th>
<th>pH</th>
<th>SOM</th>
<th>Soil RI</th>
<th>Sediment RI</th>
<th>TBA</th>
<th>HI</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SOM</td>
<td>–0.41**</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil RI</td>
<td>0.15</td>
<td>–0.24*</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sediment RI</td>
<td>–0.10</td>
<td>0.08</td>
<td>–0.1</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TBA</td>
<td>–0.03</td>
<td>0.12</td>
<td>–0.08</td>
<td>0.02</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>HI</td>
<td>–0.15</td>
<td>0.05</td>
<td>–0.13</td>
<td>0.33**</td>
<td>–0.22*</td>
<td>1</td>
</tr>
</tbody>
</table>

*P < 0.05; **P < 0.01.
calculation not only based on the contents of heavy metals in plants, but also based on the daily plants’ ingestion.

3.5. Potential uncertainties in the risk assessment

Several uncertainties affect the risk assessment performed in this study:

1. Uncertainty in classification of risk indexes’ grades. Each risk assessment has its own evaluation standard. According to the evaluation standards, risk indexes for assessment could be classified into several grades. However, binary categorization (taking values of 0 or 1) based on boundaries between grades was one of the important factors influencing the uncertainty of risk assessment (Ghaemi et al., 2014). In fact, risk indexes should be soft classified, which permits an assessment that does not require binary categorization but allows grades of risk indexes to be described for probability within the unit interval [0, 1] (Mcbratney and Odeh, 1997).

2. Uncertainty in auxiliary data of health risk assessment. The health risk assessments, which were estimated through consumption of vegetables, could lead to uncertainty in estimates (Hu et al., 2017). The sum of vegetable consumption was adopted based on data from the Exposure Factors Handbook of Chinese Population issued by the China National Environmental Protection Agency (CNEPA, 2013). It was assumed that the consumption of leafy, rootstalk and fruit vegetables is equal, which could lead to uncertainty in the estimates. With the rapid development of transportation in China, the sources of vegetables have become diversified; complex vegetable sources could influence vegetable consumption, leading to uncertainty in health risk assessments.

3. Uncertainty in preferences of geodetector model. This study was conducted under specific conditions for a specific area. The selection of impact factors (independent variables) was based on knowledge from previous studies in the region (Dong et al., 2015; Hu et al., 2018), and on exploratory data analysis to estimate factors’ grid size and classification. If the classifications of model parameters are changed, the extents of model outputs reacting to HI might also be different.

4. Conclusions

An integrated assessment of multiple environmental media and was used to infer the relative influence of heavy metal factors that affect health risk in the Yangtze River basin of Nanjing, Southeast China. Cadmium displayed the highest potential ecological risk in soils and sediments, as it possessed high BA and BCF. The sediments along the Yangtze River represented the significantly highest risk in all sites. Rice was identified as the plant with the significantly highest health risk.

The determinant power of the above factors influencing health risk indicated that plant types has a highest effect on the levels of risk to human health, followed by the SOM and bioaccessibility of the heavy metal. Pollution from the upper reaches of the river and agricultural activities had a greater impact on the health risk than had industrial activities in the study area.

In view of the accumulation of Cd and the significant role of plant types and SOM, regular monitoring, source control, and integrated agricultural management should be implemented to control and reduce heavy metal inputs and improve the safety of agricultural plants. The integrated assessment concept developed and examined in this study should be considered to address other environmental problems on a regional scale. Finally, the geodetector model approach with minor adjustments was found to be intuitive and accurate for the purposes of analyzing environmental contaminants.

Fig. 6. Soil profile distribution of pH, SOM, and heavy metals in the study area.

Fig. 7. Determinant power of factors influencing hazard index for health risk.
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Appendix A. Supplementary data

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References


