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# Concentrations of Heavy Metals in Suburban Horticultural Soils and Their Uptake by *Artemisia selengensis*

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## ABSTRACT

A total of 222 surface soil samples and 40 plant samples were collected to investigate the spatial distribution and possible sources of soil heavy metals and to know the uptake and translocation of heavy metals from roots to different plant parts in a representative vegetable production area in the Baguazhou Island, a suburb of Nanjing City, East China. The arithmetic mean values of total Cd, Cr, Cu, Ni, Pb, and Zn concentrations in the soils were 0.314, 133, 41.0, 58.0, 31.8, and 114 mg kg$^{-1}$, respectively. All of these values were above the topsoil background values in the Nanjing area. Multivariate and geostatistical analyses showed that soil Cd contamination was derived mainly from agricultural practices. In contrast, Cu and Zn were derived mainly from soil parent materials and Pb from atmospheric deposition from highway gasoline stations. *Artemisia selengensis*, a locally important specialty vegetable, accumulated heavy metals primarily in the edible leaves. The general distribution of heavy metal concentrations in this plant species showed that the highest occurred in the leaves, intermediate in the stems and lowest in the roots. Cd had the highest concentration factor (root-to-soil ratio) and may pose increased health risks in the future to the local population through the consumption of contaminated vegetables.

*Key Words*: concentration factor, plant uptake, pollution source, spatial distribution, translocation factor, vegetable

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Heavy metal pollution may lead to the functional disorder of agricultural and horticultural soils, interfere with crop growth, and may even threaten human health through contamination of the food chain (Lee *et al.*, 2006; Cai *et al.*, 2010; Cai *et al.*, 2012). The main sources of heavy metals may include various anthropogenic activities such as industry, mining, atmospheric deposition, excessive application of chemical fertilizers and pesticides, and sewage and wastewater irrigation (Wilson and Pyatt, 2007; Khan *et al.*, 2008; Wang *et al.*, 2013). Heavy metals can enter plant tissues by uptake from contaminated soils, wastewater irrigation and atmospheric deposition (Huang *et al.*, 2007; Sridhara Chary *et al.*, 2008; Hani and Pazira, 2011; Wang *et al.*, 2013).

Heavy metal pollution of Chinese suburban soils has increased over the last three decades because of rapid industrialization and urbanization (Cheng, 2003; Li *et al.*, 2004; Tan *et al.*, 2006). Commercial and residential vegetable growing areas are often located in the suburbs of the cities and are subject to anthropogenic contamination such as atmospheric deposition, waste disposal, urban effluents, vehicle exhausts, fertilizer application, and long-term application of sewage sludge to agricultural land (Bilos *et al.*, 2001; Hlavay *et al.*, 2001; Koch and Rotard, 2001; Kachenko and Singh, 2006). The application of agrochemicals has increased in Baguazhou Island, a suburb of Nanjing City, East China during the past three decades in order to increase soil fertility. Some of the fertilizers and pesti-

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cides contain heavy metals such as Cd, Hg, Pb, and Zn and repeated application of these agrochemicals and other soil amendments may have increased the accumulation of heavy metals in the soil. *Artemisia selengensis* is a perennial herbaceous plant of Compositae and grows near river banks or in swamps, with wild populations widely distributed in Northeast, North and Central China. *A. selengensis* contains pharmacologically active substances including cineole, artemisia alcohol, and $\beta$-caryophyllene.

The main objectives of this study were to assess the concentrations and distribution patterns of soil heavy metals, to investigate the major sources of heavy metal pollutants, and to know the uptake and translocation of heavy metals from roots to different parts of *A. selengensis* in a representative vegetable production area in the Baguazhou Island of Nanjing City, which will provide a basis for effective targeting of policies to protect soils and consumers from long-term accumulation of heavy metals through the food chain.

## MATERIALS AND METHODS

### *Study area*

The study area, the Baguazhou Island (32°12′4.6″ N, 118°50′112.3″ E), was located in the northeast of Nanjing City, Jiangsu Province, East China (Fig. 1). The topography of Baguazhou Island is flat with a maximum elevation of 5 m. The climate is subtropical with an average annual temperature of 16.7 °C and 237 frost-free days. The average rainfall is 1 239 mm, more than 70% of which occurs from June to September. The riverine substrate is dominated by paddy soils (Agrudalfs according to the USDA Soil Taxonomic Classification) with a small quantity of fluvo-aquic soils. The study area has been traditionally associated with agricultural activities and is currently used for vegetable production. About 60% of the total field area (56 km$^2$) in the island is used for the cultivation of a special medicinal vegetable, edible wild wormwood (*Artemisia selengensis*). In the past three decades this area has been used for the continuous cropping of vegetables and large amounts of inorganic fertilizer such as urea and composite fertilizers have been used during the last two decades.

### *Sample collection and preparation*

A total of 222 soil samples were collected from the field. The sampling points were selected based on a regular grid of 400 m × 400 m and each grid had at least one sampling point (Fig. 1). Samples were collected from the top 20 cm of the soil profile. Each composite sample consisted of 3–5 sub-samples obtained using a stainless steel hand auger. All of the sampling points were recorded using a hand-held global positioning system. Soil samples were collected first from selected farms in July 2012. On the basis of the initial results additional soil samples were collected in March

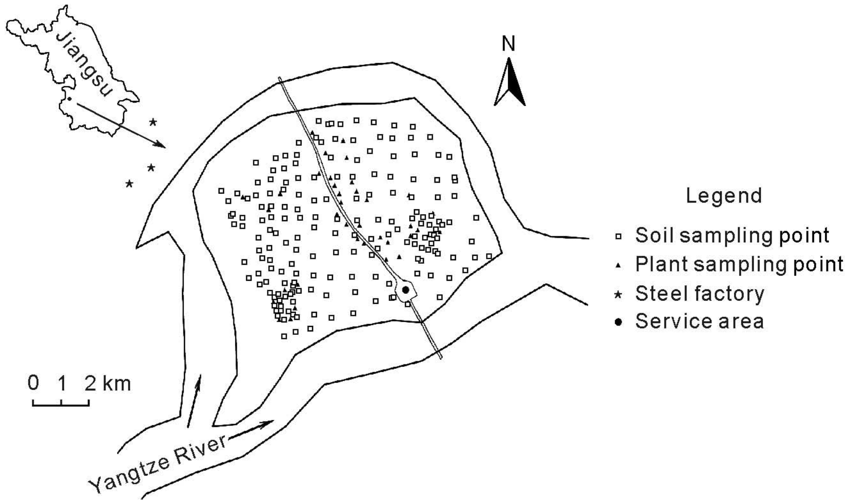

Fig. 1 Location of the study area and the sampling points in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China.

2013. All soil samples were air-dried at room temperature, stones and other debris were removed, and the samples were then passed through a 2-mm nylon sieve. Portions of soil samples (about 50 g) were ground in an agate mortar and sieved through a 0.149-mm mesh. The prepared soil samples were then stored in polyethylene bottles for analysis.

Forty vegetable samples (*A. selengensis*) were uprooted and gathered from polytunnel greenhouses at the end of harvest seasons and their corresponding soils were also sampled in March 2013. At each sampling plot, 3–5 soil subsamples were collected and combined for further treatment and chemical analysis. The fresh vegetable samples were placed in clean plastic bags and transported to the laboratory as soon as possible and then separated into roots, stem and leaves. The vegetable samples were washed with tap water to remove dust and extraneous matter and then with deionized water, dried in an oven for 72 h at 70 °C and weighed to determine their water content. The dried samples were then ground, homogenized and stored in tightly closed clean sample bottles until analysis.

### *Chemical analysis*

Soil pH was determined in an aqueous extract with a soil-to-water ratio of 1:2.5 (weight/volume) using a pH meter. The total concentrations of heavy metals were determined by inductively coupled plasma-mass spectrometry (VGPQ II Agilent 7500a, Agilent, California, USA). About 0.2 g dried soil sample was treated with 10 mL 1:1 (volume/volume) concentrated $HNO_3$ and HCl, and placed in a Teflon vessel digester at $> 120$ °C for 480 min. The acid extracts were reduced to a minimum volume on hot-plates and re-diluted with 1% $HNO_3$ for analysis. Plant materials for metal analysis were treated with 2 mL 30% $H_2O_2$ and 6 mL 69% $HNO_3$ to mineralize cellulose, and then prepared in a similar manner to the soil samples.

Quality assurance and quality control (QA/QC) for heavy metals in soil and plant samples were estimated by determining the heavy metal contents in blank and duplicate samples and certified reference materials (CRMs). The certified reference materials, including GBW07402 (chestnut colored soil), GBW07404 (calcareous soil) and GBW10014 (for plants), were obtained from the China National Center for Standard Reference Materials. Blank and duplicate samples and the CRMs were included with every 10 samples in the analysis. The elemental recoveries and relative standard deviation for the CRMs were 92%–108% and $<$ 3.6%, respectively.

### *Data analysis*

Pearson's correlation coefficient analysis and principal component analysis (PCA) were performed (Facchinelli *et al.*, 2001; Zhang *et al.*, 2008), using the commercial statistical software package SPSS version 17.0 for Windows, to identify the relationships among heavy metals in the soils and their possible sources. The correlation coefficient measured the strength of the inter-relationship between two heavy metals. PCA, a multivariate analytical tool, was used to reduce a set of original variables to a small number of latent factors (principal components) and analyze the relationships among the heavy metals. In addition, the normal distribution of each metal's concentration was evaluated using the Kolmogorov-Smirnov test based on the raw data.

Geostatistics provides a set of statistical tools for incorporating the spatial and temporal coordinates of observations in data processing (Saito and Goovaerts, 2000). Kriging, a geostatistical interpolation method, uses the semivariogram to quantify the spatial variability of regionalized variables and provides parameters for spatial interpolation. The maps of the spatial distribution of heavy metal concentrations were generated by kriging interpolation with the support of ArcGIS-Geostatistical Analyst software.

The concentration factor (CF) is defined as the ratio of the heavy metal concentrations in roots to those in the soil. The translocation factor (TF) is the ratio of the heavy metal concentrations between one part and another part of the plant (*i.e.*, between stem and root or between leaf and stem), which can characterize the difficulty of heavy metal translocation in the plant system (Kloke *et al.*, 1984; Yang *et al.*, 2008).

## RESULTS AND DISCUSSION

### *Soil heavy metal concentrations and distribution characteristics*

Soil pH values changed greatly from 3.87 to 8.20

and 41.4% of the soils had pH above 7.5, 32.9% between 6.5 and 7.5, and only 25.7% below 6.5. This may be partly ascribed to the parent materials, topography, soil texture, and vegetation (Helyar *et al.*, 1990; Barton *et al.*, 1994; Álvarez *et al.*, 2002), and also resulted from the long-term application of chemical fertilizers (Berg, 1986; Schwab *et al.*, 1990; Conyers *et al.*, 1996; Wei *et al.*, 2006; Liao *et al.*, 2007).

Table I presents the descriptive statistics of heavy metal concentrations in the horticultural top soils and the background values of Nanjing soils. There was a distinct change in the concentrations of heavy metals among the soil samples and the concentrations of Cd, Cr, Cu, Ni, Pb and Zn varied from 0.101 to 0.513, 43.1 to 590, 14.7 to 85.2, 19 to 291, 13.2 to 59.7, and 43 to 216 mg kg$^{-1}$, respectively. The mean values of the soil heavy metal concentrations followed the descending order of Cr > Zn > Ni > Cu > Pb > Cd and were all higher than their background values (1.65, 2.26, 1.27, 1.66, 1.28, and 1.49 times the corresponding background values, respectively), clearly demonstrating an anthropogenic contribution. However, except for Cd and Ni, the values of Cr, Zn, Cu, and Zn were lower than the "Grade II" limit of the Chinese Environmental Quality Standard for Agricultural Soils (GB-15618-1995). The coefficients of variation varied from 18.2% for Zn to 111% for Cr and decreased in the order of Cr > Ni > Pb > Cu > Cd > Zn, indicating the significant influence of an anthropogenic contribution to soil Cr and Ni concentrations such as industrial pollution (Wu *et al.*, 2011; Hernández-Quiroz *et al.*, 2012) and vehicle traffic (Wu *et al.*, 2011). Large standard deviations were found for all heavy metals except Cd. This also indicated the wide variation in metal concentrations in the horticultural soils. The K-S test confirmed that the concentrations of Cr and Ni in the soils were not distributed normally, showing positively skewed data. The main contribution to heavy metal contamination in the soils of the Baguazhou Island was due to Cr and Ni and accumulation of Cd, Zn, and Cu concentrations in the soils was also found. In accordance with the Environmental Quality Standard for Agricultural Soils (GB15618-1995), the mean concentrations of Cd, Zn, Cu, Pb and Cr in the top soils were lower than the maximum allowable concentrations in Chinese agricultural soils with the exception of Ni. The percentages of sampled soils exceeding the maximum allowable concentrations were 11% for Cd, 8% for Cu, 14% for Cr and 41% for Ni. This indicated that local anthropogenic activities posed a threat to food chain safety by vegetable production.

Soil pH is generally regarded as the most important factor affecting the solubility of heavy metals and their availability to plants (Reddy and Patrick, 1977; Zhao *et al.*, 2010; Zeng *et al.*, 2011). When soil pH decreased, pronounced increases of heavy metal mobility and bioavailability were observed (Badawy *et al.*, 2002; Wang *et al.*, 2006), thus enhancement of the uptake of heavy metals by plants and thereby the threat to human health need be concerned (Brallier *et al.*, 1996; Oliver *et al.*, 1996). Soil acidification can, therefore, be an effective method to promote the uptake of heavy metals by plants and then minimize the toxic effects. However, the higher mobility resulting from soil pH

TABLE I

Basic descriptive statistics$^{a)}$ of soil heavy metal concentrations in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China

| Heavy metal | Mean | Maximum | Minimum | Median | SD | Skewness | Kurtosis | CV | K-S statistic | Background value | EQS$^{b)}$ Grade II |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | mg kg$^{-1}$ | | | | | | | % | | mg kg$^{-1}$ | |
| Cd | 0.314 | 0.513 | 0.101 | 0.316 | 0.069 | −0.005 | 0.08 | 22.0 | 0.551 | 0.19 | 0.30 |
| Cr | 133 | 590 | 43 | 103 | 156 | 7.53 | 80.0 | 111.0 | 3.94 | 59 | 200 |
| Cu | 41.0 | 85.2 | 14.7 | 41.9 | 9.33 | 0.166 | 1.76 | 22.8 | 1.01 | 32.2 | 100 |
| Ni | 58 | 291 | 19 | 48 | 35.7 | 3.00 | 11.8 | 61.5 | 3.74 | 35.0 | 50 |
| Pb | 31.8 | 59.7 | 13.2 | 31.7 | 7.38 | 0.395 | 0.63 | 23.2 | 0.629 | 24.8 | 300 |
| Zn | 114 | 216 | 43 | 116 | 20.9 | 0.124 | 2.46 | 18.2 | 1.04 | 76.8 | 250 |

a) SD = standard deviation; CV = coefficient of variation; K-S = Kolmogorov-Smirnov test.

b) Chinese Environmental Quality Standard for Agricultural Soils (GB 15618-1995).

TABLE II

Pearson correlation coefficients among soil heavy metal concentrations in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China

| Heavy metal | Cd | Cr | Cu | Ni | Pb | Zn |
|---|---|---|---|---|---|---|
| Cd | 1 | | | | | |
| Cr | −0.060 | 1 | | | | |
| Cu | 0.400** | 0.039 | 1 | | | |
| Ni | 0.202** | 0.485** | 0.136* | 1 | | |
| Pb | 0.064 | 0.033 | 0.701** | 0.033 | 1 | |
| Zn | 0.518** | 0.034 | 0.703** | 0.131 | 0.494** | 1 |

*, **Significant at $P < 0.05$ and $P < 0.01$, respectively (2-tailed).

decrease could also imply heavy metal displacement to deeper soil layers and higher risk of water system contamination by leaching or runoff of heavy metals.

### *Correlations among heavy metal concentrations*

Correlation analysis provides an effective way to reveal the relationships among multiple variables and thus contributes to an understanding of the drivers and the sources of chemical components. Heavy metals in soils usually have complicated inter-relationships. Significant positive correlation between soil heavy metals may reflect similar pollution sources (Micó *et al.*, 2006; Li *et al.*, 2013). The calculated Pearson correlation coefficients and their significance levels are shown in Table II. The concentration of Cd showed highly significant positive relationships with Cu (0.400), Ni (0.202), and Zn (0.518). In addition, the correlations between Cu and Pb and Zn were significant and positive ($P < 0.01$). However, the concentrations of Cr and Ni showed weak correlations with Pb and Zn, indicating that Cr and Ni may have been derived from different sources from that Pb or Zn.

### *Principal component analysis of soil heavy metals*

Principal component analysis (PCA) was adopted to identify the origin of the soil heavy metals (Table III). The eigen-values of the three extracted components were all greater than 1.0. The heavy metals were, therefore, grouped into a three-component model that accounted for 88% of the total variance in the data. The component matrix (Table IV) showed that Cu, Pb, and Zn were strongly associated with the first principal component (PC 1) which explained 43.3% of the total variance, while Cr and Ni were mainly distributed with the second principal component (PC 2) which explained 27.6% of the total variance. The third principal component (PC 3) was correlated very strongly with Cd which had a high loading value (0.757), accounting for 17.0% of the total variance. The metals in PC 1 were mainly derived from non-anthropogenic sources, indicating local natural sources. The most important sources of Cd in the arable soils of suburban areas are the long-term application of fertilizers, sewage sludges, and organic manures (Zhang, 2006; Hani and Pazira, 2011). Cr and Ni in PC 2 were strongly correlated with each other and were clearly separated from the other

TABLE III

Principal component analysis for soil heavy metal concentrations in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China

| Principal component | Eigen-value | Proportion of total variance | Cumulative proportion |
|---|---|---|---|
| | | % | |
| 1 | 2.601 | 43.342 | 43.342 |
| 2 | 1.659 | 27.646 | 70.989 |
| 3 | 1.020 | 17.007 | 87.996 |
| 4 | 0.320 | 5.328 | 93.324 |
| 5 | 0.221 | 3.684 | 97.008 |
| 6 | 0.180 | 2.992 | 100.000 |

TABLE IV

Principal component (PC) matrices correlated with soil heavy metal concentrations in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China

| Heavy metal | PC 1 | PC 2 | PC 3 |
|---|---|---|---|
| Cd | 0.564 | −0.150 | 0.757 |
| Cr | 0.357 | 0.869 | −0.168 |
| Cu | 0.883 | −0.385 | 0.059 |
| Ni | 0.399 | 0.849 | 0.171 |
| Pb | 0.715 | −0.194 | −0.587 |
| Zn | 0.840 | −0.256 | 0.145 |

heavy metals regarding their correlation coefficient analysis and PCA. This suggested that Cr and Ni were derived mainly from anthropogenic sources such as industrial activities and traffic pollution.

*Spatial distribution of heavy metals*

The spatial distribution of heavy metals is a useful aid to assess the possible sources of enrichment and to identify hotspots with high metal concentrations. The estimated concentration of Cd, Cr, Cu, Ni, Pb and Zn are presented in the following geochemical maps (Fig. 2), which identified several hotspots with high metal concentrations.

The hotspots where the soils were enriched with Pb mostly coincided with highway gasoline stations with high vehicle exhausts. Soils act as a sink for Pb from atmospheric deposition. Pb aerosols can be transported over long distances in the atmosphere, resulting in high concentration of Pb in the soils around the pollution sources. However, the transport of heavy metals from urban to surrounding areas is limited to a circumference of approximately 1 km, with Pb concentrations showing a declining trend with increasing distance from the pollution source (Biasioli *et al.*, 2006).

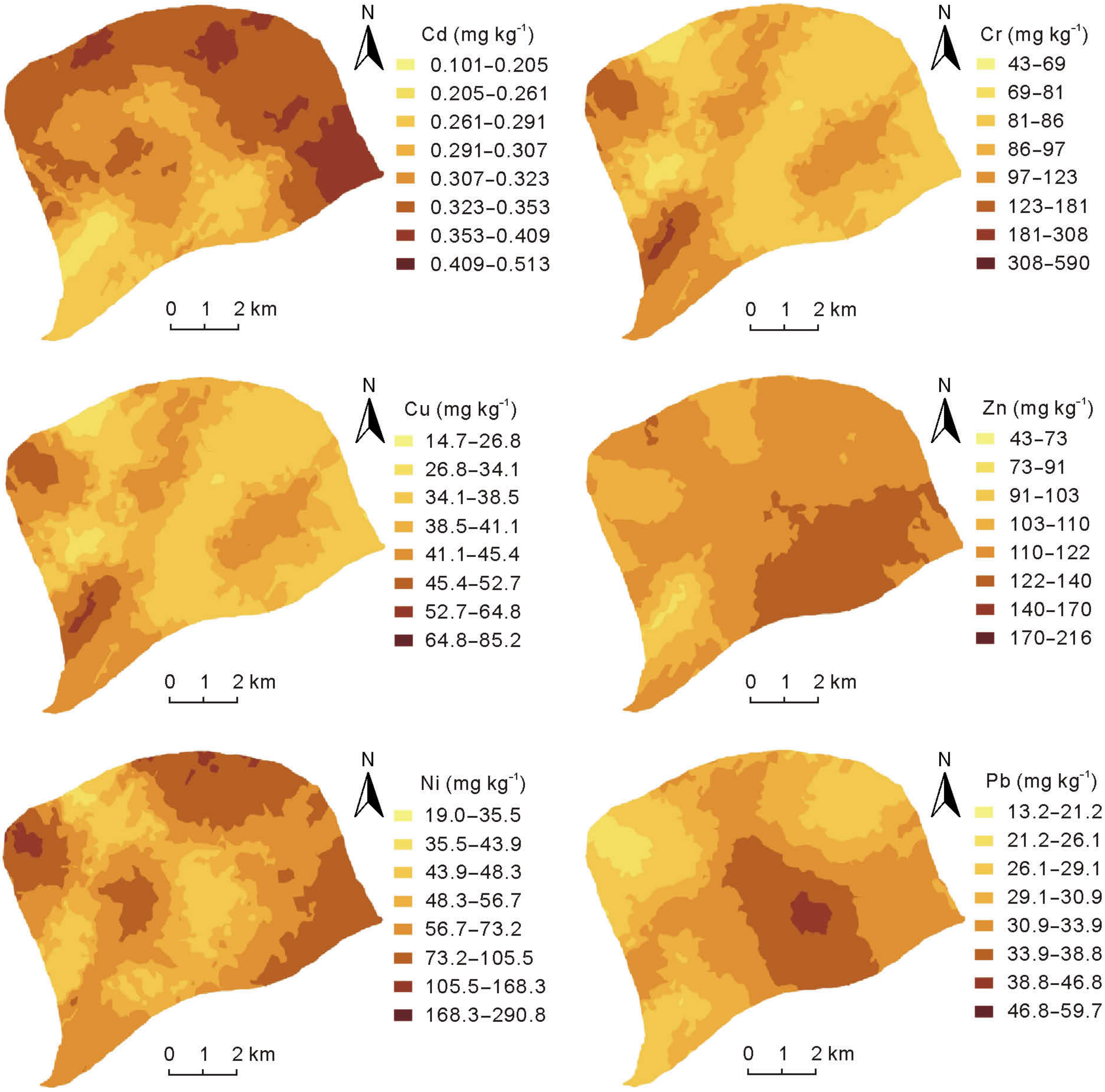

Fig. 2 Spatial distribution of soil heavy metal concentrations in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China.

The higher Cd concentration tended to occur in the southeast and northeast and this may be associated with agricultural activities such as application of pesticides, phosphorus fertilizers and plastic film (for plastic greenhouses or mulching) containing Cd (Kachenko and Singh, 2006; Montagne *et al.*, 2007).

The concentrations of soil Cu and Zn were higher in the southeast of the study area and their spatial distribution showed a good correlation with the distribution of riverine and marine deposits, indicating the influence of soil parent materials.

Another pollution hotspot was evident at the northwest corner of the study area. This area was most polluted with Cr and Ni and second most polluted with Cd. Many heavy industries such as steelworks and chemical industries are located nearby across the river and this pollution hotspot may, therefore, reflect industrial inputs.

### *Distribution of heavy metals in plant parts*

The concentrations of 6 heavy metals in the *A. selengensis* samples (Fig. 3) were all below the Chinese maximum allowable concentrations in food (GB2762-2012: Pb 0.1 mg kg$^{-1}$, Cd 0.1 mg kg$^{-1}$, Cr 0.5 mg kg$^{-1}$, and Ni 1.0 mg kg$^{-1}$; GB15199-94: Cu 10 mg kg$^{-1}$; and GB13106-91: Zn 20 mg kg$^{-1}$). The concentrations of Cd, Pb, and Zn in different plant parts showed the order of leaves > stems > roots. The leaves were the main site of heavy metal accumulation. However, Cu concentrations followed the sequence of stems > leaves > roots. The tissue concentrations of Ni showed the order of stems > roots > leaves and Cr had a similar trend. The relatively low concentrations of Cd, Pb, and Zn in the roots and stems may be due to the transport function of these plant parts. It can also be indicated that *A. selengensis* might absorb heavy metals from the air or by atmospheric deposition through its leaves (Luo *et al.*, 2011), resulting in higher concentrations of heavy metals in the leaves than in the roots and stems. In addition, the differences in the accumulation and distribution of heavy metals among the different plant parts may be related to the differences in the characteristics of heavy metals, their toxicity, and their concentrations in the soils.

The concentration factors (CF) and translocation factors (TF) were calculated to study the accumulation and transport characteristics of heavy metals in different plant parts (Kloke *et al.*, 1984; Yang *et al.*, 2008). In general, heavy metals showed distinct differences in their concentration factors. The concentration factor of Cd was the highest and the CFs of Cd in the leaves, stems and roots were 1.80, 1.57 and 1.08, respectively, indicating that the plants had very strong Cd enrichment capacity. The plant CF of Cu was the second highest, with values in the leaves, stems and roots of 0.781, 0.783 and 0.604, respectively. Generally, Cu and Zn are taken up as micronutrients essential for plant growth. However, Pb and Cr are nonessential and their root uptake was limited as their diminished transfer capacities from roots to stems and leaves (0.003 and 0.014, respectively).

Table V shows the TFs of heavy metals in the plants and the values followed the order of Pb > Cd > Zn > Cu > Ni > Cr. This agrees with the results of some previous studies with the sole exception of Pb (Khan *et al.*, 2008; Zhuang *et al.*, 2009; Cao *et al.*, 2010). As a result, Pb may have been readily transported from roots to stems but Ni and Cr were relatively immobile. From stems to leaves the TF of Pb was the highest at 2.05 and that of Ni was the lowest at 0.926. The translocation ability of each plant part indicates some resistance to related heavy metals. The higher the translocation ability for heavy metals of a tissue is, the lower the resistance and the lower its retention capability are (Yang *et al.*, 2008).

## CONCLUSIONS

Anthropogenic activities affected the heavy metal concentrations in the soils and vegetables in the Baguazhou Island, but most of the soil samples were still safe for vegetable production according to the Chinese Environmental Quality Standard for Agricultural Soils. Soil Cu and Zn were derived mainly from the soil parent materials. Local contamination from agricultural practices, particularly the application of fertilizers and manures, was the primary source of Cd contamination in the soils, and atmospheric deposition played an important role in soil Pb accumulation. Cr and Ni had anthropogenic origins and their spatial distributions showed a point source contamination, suggesting significant anthropogenic inputs of Cr and Ni in the study area. The locally important speciality vegetable

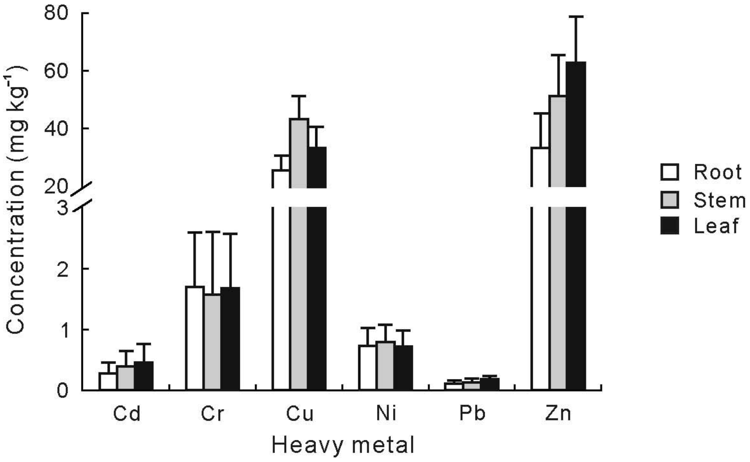

Fig. 3 Concentrations of heavy metals (dry weight basis) in roots, stems, and leaves of *Artemisia selengensis*, a locally important speciality vegetable in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China.

TABLE V

Translocation factors of heavy metals in *Artemisia selengensis*, a locally important speciality vegetable in the Baguazhou Island, a suburb of Nanjing City, Jiangsu Province, East China

| Translocation factor | Cd | Cr | Cu | Ni | Pb | Zn |
|---|---|---|---|---|---|---|
| Between stem and root | 1.663 | 1.095 | 1.341 | 1.177 | 1.830 | 1.657 |
| Between leaf and stem | 1.296 | 1.236 | 1.000 | 0.926 | 2.045 | 1.239 |

*A. selengensis* had the highest CF for Cd and the second highest for Cu. Plant uptake capacity for Pb and Cr was relatively low. Cd showed the highest CF and TF in *A. selengensis*, indicating that this plant species had a high capacity for Cd uptake. These results may provide a basis for the effective targeting of policies to reduce metal inputs and to protect soils and consumers from long-term heavy metal toxicity.

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