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Temporal variations and spatial distributions of heavy metals in a wastewater-irrigated soil-eggplant system and associated influencing factors

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ABSTRACT

Heavy metal pollution in farmlands is highly concerned as crops' easy-uptake of heavy metal can ultimately affect consumers. In order to offer suggestions on cultivating safe quality vegetable, specifically eggplant which is widely consumed for its nutritional value and antioxidant activity, a field study was undertaken to investigate the temporal variations and spatial distributions of heavy metals in a wastewater-irrigated soil-eggplant system. In the present study, eggplants were planted in the farmlands of Weichuan village (WC) (relatively unpolluted field), Liangzhuang village (LZ) (moderately polluted field) and Minqin village (MQ) (seriously polluted field) to elucidate their temporal uptake processes of heavy metals described by the sigmoid model. Eggplant tissues from severely polluted farmlands were found with higher heavy metal concentrations and lower yields compared with other two groups. What is more, 25 farmlands along the Dongdagou stream (heavy metals polluted stream) were chosen to analyze the spatial distribution of heavy metals in soils and eggplants. Heavy metal concentrations in eggplants decreased with the decline of heavy metal concentrations in soil from upstream (pollution source) to downstream. Moreover, several methods were employed to assess bioavailability of heavy metals in soils. All the bioavailable heavy metals were found in linear positive correlations with heavy metal concentrations. Meanwhile, linear correlations were found between heavy metals in soils and eggplants. At last, redundancy analysis was used to investigate the effects of soil properties (pH, organic matter and texture of soils) and heavy metals on eggplants' uptake. The results indicated that soil heavy metals had a dominant impact on their accumulations in eggplant fruit, with a variance contribution of 78.0%, while soil properties had a regulatory effect, with a variance contribution of 5.2%.

1. Introduction

Elevated heavy metal concentrations in agroecosystem derived from fertilizer application, pesticide spraying and wastewater irrigation is of growing concern (Khan et al., 2013; Kupper and Andresen, 2016; Lu et al., 2015). Vegetable consumption is one of the major pathways of human exposure to heavy metals (Xu et al., 2013). Heavy metals can pose a series of harmful damages to plants as well as consumers. For instance, lead (Pb) exposure may cause various morphological, physiological and biochemical dysfunctions in plants and lead to plumbism and anemia to human (Shahid et al., 2011; Zukowska and Biziuk, 2008). Whereas, cadmium (Cd) exposure can result in vegetables' decreasing growth and biomass (Gharaibeh et al., 2015; Kumar et al., 2016), and lead to renal dysfunctions via vegetables consumption (Cui et al., 2005). Although copper (Cu) and zinc (Zn) are essential elements for many physiological processes in plants, they can yet become very toxic at elevated levels (Anjum et al., 2015; Nagajyoti et al., 2010).

Eggplant (*Solanum melongena* L.), a popular vegetable, is widely cultivated for its fruits containing phytochemicals and nutraceuticals and rich in nutritions, which are well-known for their antioxidant activities and healing effects on certain diseases (Azuma et al., 2008; Friedman, 2015; Kwon et al., 2008). Recent studies suggested that safety of eggplants consumption from polluted farmlands has suffered because of elevated levels of heavy metals such as Pb and Cd accumulation in eggplant (Saeedifar et al., 2014; Ye et al., 2015). However, only few studies have described the temporal variations and spatial distributions of heavy metal accumulation in eggplant, which could offer suggestions for cultivating eggplant of safe quality.

In previous studies, heavy metals in plants were investigated in pot and field experiments to evaluate their temporal variations (Lai and Chen, 2013; Zhu et al., 2016). What is more, the dynamic increase of plant biomass and the temporal variations of heavy metal uptake by

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plants were found to follow a sigmoid model (Molina et al., 2013). Moreover, the sigmoid model was proved to be able to simulate the processes of Chinese cabbage and radish's dynamic growth and heavy metal uptake (Ai et al., 2016). Therefore, sigmoid model was utilized to describe the dynamic growth and heavy metal uptake of eggplant in the present study. On the other hand, many studies described the spatial distribution of heavy metals in soils and sediments using the geographical information science (GIS)-based approach which was regarded as a powerful tool to survey heavy metal pollution (Hou et al., 2017; Liu et al., 2016). Hence, ArcGIS 10.3 was employed to construct the spatial distribution maps of heavy metals in soils and eggplants in the present study.

Bioavailable metals in soil, rather than the total ones, are actually responsible for the uptake, accumulation and redistribution of heavy metals in crops, since total heavy metal concentration in soil may not necessarily reflect the uptake of heavy metals by crops (Liu et al., 2015; Zeng et al., 2011). Furthermore, soil properties such as pH, organic matter and soil texture are key factors in influencing heavy metal bioavailability in soil (Chen et al., 2016; Kirkham, 2006). A negative correlation was found between soil pH and bioavailable Cd and Cd uptake increased under lower soil pH (Bilodeau-Gauthier et al., 2011; Shahid et al., 2011). Soil organic matter has an immobilisation effect on heavy metals in soils which results in decreasing Pb uptake in plants, whereas, clay can effectively influence heavy metal bioavailability through specific adsorption and cation exchanges (Rieuwerts et al., 2006; Shaheen et al., 2013; Yan et al., 2017). In addition, many analytical methods were efficiently used to clarify the correlations between soil properties and heavy metal uptake by plants (Xiao et al., 2017; Yang et al., 2016). Therefore, Pearson correlation, multivariate regression analysis and redundancy analysis (RDA) were utilized to investigate the effects of soil properties on heavy metal uptake by eggplant in the present study.

Herein, the present study intended to (1) clarify the temporal variations of heavy metals in eggplant (the dynamic process of heavy metal uptake by eggplant); (2) investigate the spatial distribution of heavy metals in soils and eggplants along with a polluted stream; (3) discuss the correlation between heavy metals in soils and eggplants; (4) analyze the effects of soil properties and heavy metal concentrations on the heavy metal uptake by eggplant. The present study attempts to provide suggestions for cultivating safe quality vegetable by exploring the temporal variations and spatial distributions of heavy metals in a wastewater-irrigated soil-eggplant system and the associated influencing factors.

2. Materials and methods

2.1. Study area

A battery of farmlands (36.4313002°–36.5395874° N, 104.1964310°–104.3956182°E) were chosed to plant eggplants in Baiyin city along the Dongdagou stream which originates from the local industrial and mining districts and flows into the Yellow River making these farmlands polluted with heavy metals in different degrees (Fig. 1). These farmlands are located in the narrow belt-like valley and are being irrigated with wastewater from the upstream for last couple of decades. As is reported, Cu, Zn, Pb and Cd are found at high concentrations in soils and crops, thus are deemed as the main pollutants in the farmland soils (Liu et al., 2016; Nan and Zhao, 2000). As a control, a farmland (35.9455306°N, 103.2552278°E) in Liujiaxia district (a famous scenic spot in Gansu) was chosen as the non-polluted reference site.

2.2. Field experiment

Dynamic growth experiment was carried out in 2015 to clarify the temporal variations of heavy metals in eggplant. A farmland in Weichuan village (WC) (Liujiaxia district) was taken as the non-polluted reference site. Whereas two farmlands from Liangzhuang village (LZ) and Minqin village (MQ) (Baiyin city) were chosen as the moderately and seriously polluted sites respectively. At sowing time, eggplant seeds from the same stock were sowed in holes (3–5 seeds per hole) in three quadrats of 15 m × 10 m with 30 cm × 50 cm row spacing. The plant density was more than 200, 000 plants/ha at sowing time. Then, thinning out of seedlings was done when the seedlings were with two true leaves. At harvest time, plants densities of 65, 400 plants/ha for WC, 67, 500 plants/ha for LZ and 63, 800 plants/ha for MQ were noted. The growth period lasted from May. 7th to Sep. 5th in 2015. After germination, 5–10 whole plants were randomly collected every 14 days until 112th day. Fruit period was noted from 56th day to 112th day. During the period of eggplant sampling, corresponding topsoil samples (0–20 cm depth) were also collected from the same fields.

Spatial distribution experiment was conducted in August 2016. Twenty-five sites (S1 to S25) along the Dongdagou stream were chosen to collect samples of soils and eggplants. When sampling, five subsamples were mixed into one composite sample. From each site, one paired composite samples of soil (0–20 cm depth) and eggplant were randomly collected at harvest time for further analyses. All analyses were performed in triplicate for every composite sample.

2.3. Plant and soil analysis

In the laboratory, tissues of eggplants (root, stem, leaf and fruit) were separated and washed with tap water followed by being washed with deionized water. After being oven-dried at 70 °C for 48 h, all the plant samples were ground into powder with a grinder. The fresh and dry weights of samples were recorded using an electronic balance. Soil samples were air-dried and ground to pass through 0.2 mm nylon sieves. Soil pH was measured using a pH meter in the mixture of soil and deionized water (1:2.5, w/v). Organic matter content was determined by the method of potassium dichromate-dilution heat titration (Walkley and Black, 1934). Soil texture (clay, silt, and sand) was measured by a laser particle size analyzer (Mastersizer 2000, Malvern Instruments Ltd., UK). To determine heavy metal concentrations in soils and eggplant tissues, soil samples were digested with a mixture of HCl, HNO_3, $HClO_4$ and HF, while eggplant samples were dealt with HNO_3 and $HClO_4$ (Ai et al., 2016).

Reagent blanks were run parallel to samples in order to validate our results. Certified biological (GBW 10052, GSB - 30) and soil (GBW 07402, GSS - 2) reference materials were employed to ensure quality of experiments. The results showed relative standard deviation (% RSD) was within 5% and the recovery percentages were 90–110%.

To assess the bioavailable of heavy metals in soil, three different one-step extraction methods were applied in the present study: (1) extraction with a mixture extractant (0.005 mol/L diethylene triamine pentacetate acid (DTPA), 0.100 mol/L triethanolamine (TEA) and 0.010 mol/L $CaCl_2$, pH 7.3, 1:10 w/v); (2) extraction with 0.050 mol/L ethylene diamine tetraacetic acid (EDTA) (1:10 w/v); (3) extraction with 0.100 mol/L hydrochloric acid (HCl) (1:10 w/v) (Baceva et al., 2014; Rivera et al., 2016; Xiao et al., 2015).

In addition, a sequential extraction procedure according to the European community bureau of reference (the extraction procedure was defined as BCR method in the present study) was used to analyze the chemical speciation of heavy metals in soil (Ure et al., 1993). The three extracted fractions were defined as BCR_1, BCR_2 and BCR_3. The procedure is briefly described as follows:

Step 1: acid-soluble/exchangeable fraction (BCR_1). Extraction of soil sample was done with 0.11 mol/L CH_3COOH (1:40 w/v, shaking for 16 h at 20 ± 2 °C).

Step 2: reducible fraction (BCR_2). Residues from step 1 were shaken with 0.1 mol/L NH_2OH•HCl (adjust to pH 2.0 with HNO_3, 1:40 w/v, shaking for 16 h at 20 ± 2 °C).

Step 3: oxidizable fraction (BCR_3). Residues from step 2 were digested with 8.8 mol/L H_2O_2 (1:10 w/v) at room temperature for 1 h.

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