Chemosphere 103 (2014) 121–130

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

The impact of greenhouse vegetable farming duration and soil types on phytoavailability of heavy metals and their health risk in eastern China

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HIGHLIGHTS

- Greenhouse farming lowered soil pH and increased soil metal availability over time.
- Greenhouse farming caused greater soil pH decrease in Anthrosols than in Cambosols.
- Decreased soil pH and elevated metal availability increased metal phytoavailability.
- Increased soil OM over time had effect on crop-depending Cu and Zn uptakes.
- Potential health risks were mainly posed by Cd in leaf vegetables in selected sites.

ARTICLE INFO

Article history:
Received 26 July 2013
Received in revised form 17 November 2013
Accepted 20 November 2013
Available online 11 December 2013

Keywords:
Greenhouse vegetable production duration
Soil types
Phytoavailability of heavy metals
Potential health risks
Eastern China

ABSTRACT

Heavy metal contamination in vegetables from greenhouse vegetable production (GVP) in China requires major attention. For GVP sustainability at a large regional level, 441 surface GVP soil and 132 corresponding greenhouse vegetable samples were collected from six typical GVP bases in eastern China to systematically evaluate the impact of GVP duration and soil types (Anthrosols and Cambosols) on phytoavailability of four major metals, Cd, Cu, Zn, and Pb, and their health risk. The results revealed high Cd accumulation in leaf vegetables grown in Anthrosols, which might pose potential health risk. Regardless of soil types in the study region, greenhouse farming lowered soil pH and enhanced metal availability with rising GVP duration, which might exacerbate Cd phytoavailability and vegetable Cd contamination as well as potential health risk. Also, increased GVP soil organic matter contents over time, found in some locations, affected crop-depending Cu and Zn uptakes. Furthermore, due to GVP, the annual decrease rate of soil pH and increase rates of soil available metal concentrations were generally much greater in Anthrosols than those in Cambosols, which contributed a lot to high Cd uptake by leaf vegetables grown in Anthrosols and their potential health risk. From sustainable GVP perspective, fertilization strategy with reduced frequency and rate is especially important and effective for abating soil and vegetable contamination by heavy metals under greenhouse farming.

1. Introduction

As an intensive form of agriculture, greenhouse vegetable production (GVP) prolongs the growing season in contrast with conventional vegetable cultivation (Jensen and Malter, 1995) to provide people off-season and sufficient vegetables. Owing to market demands and economic incentives, greenhouse vegetable production has rapidly expanded for recent 20 years in many developing countries such as China (Liu et al., 2008; Huang and Jin, 2008), especially in eastern China where the majority of China's arable land is concentrated. However, to achieve high crop production, the strategy with heavy fertilization driven production has been adopted under GVP conditions in some areas in China such as North China Plain (Hu et al., 2012; Yu et al., 2010), which has linked GVP with accumulation of heavy metals, particularly Cd, Cu, Zn, and Pb in GVP soil (GVPS) (Huang and Jin, 2008; Liu et al., 2011; Yang et al., 2013).

These contaminated soils may elevate metal phytoavailability relative to uncontaminated soils (Sharma et al., 2007). The excessive ingestion of the essential elements, such as Cu and Zn, which act as micro-nutrients to maintain normal body function at low concentrations, may affect human health (Powers et al., 2003). Moreover, the ingestion of nonessential elements, such as Cd and

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http://dx.doi.org/10.1016/j.chemosphere.2013.11.047

Pb, will cause deleterious effects on human health (Jarup, 2003). Similarly, the irrigation water containing heavy metals can cross-contaminate crops grown in a soil never previously polluted with metals under greenhouse conditions (Stasinos and Zabetakis, 2013). However, the inputs of Cd, Cu, Pb, and Zn to the greenhouse vegetable plots through irrigation water were generally limited in eastern China (Yang et al., 2013), which had negligible effects on metal uptakes.

Since greenhouse vegetable consumption has become an important part of human diet, especially in China, the security associated with greenhouse vegetable production and consumption deserves great concern. Thus, it is necessary to systematically assess heavy metal uptake by greenhouse vegetables and subsequent health risk. In contrast with such an assessment, a comprehensive investigation of how phyto-availability of heavy metals, especially Cd, Cu, Zn, and Pb, and their health risk change with the development of GVP is more important for providing preliminary data for developing appropriate strategies of controlling heavy metals in this agricultural system and further facilitating sustainable GVP. However, such a study is lacking. As is well known, the metal phyto-availability is controlled by soil pH and organic matter content (Zeng et al., 2011). Soil total and available metals are also of great importance to predict metal phyto-availability (Brun et al., 1998; McLaughlin et al., 2000). Nevertheless, greenhouse vegetable farming duration may cause the drastic change of these soil parameters. Given the wide and rapid development of GVP in eastern China, the soil types of this region are also important factors influencing the change extent of these soil parameters.

Therefore, the objectives of this research were to (i) investigate pH, OM and present accumulation status of Cd, Cu, Zn, and Pb in GVPS and greenhouse vegetables in eastern China, (ii) discuss the detailed changes of these soil parameters related to greenhouse vegetable farming duration and the change extent among different soil types, (iii) estimate metal phytoavailability with these soil parameters and assess their potential health risks and (iv) conclude the specific impacts of GVP duration and soil types on metal phytoavailability and their health risks based on the changes of soil parameters.

2. Materials and methods

2.1. Study area and fertilization description

Since the majority of China's arable land and most greenhouse vegetable production (GVP) in China are concentrated in the east, this research was conducted in this region. Three typical areas were selected, including Nanjing City, the capital of Jiangsu Province, Tongshan County in Jiangsu Province and Shouguang County in Shandong Province. Topographically the study areas in Nanjing are gently undulating and soils are primarily comprised of Stagnic Anthrosols (Gong et al., 2003) derived from loess parent material with clay loam to clay texture. While the greenhouse plots in Tongshan and Shouguang had flat elevation and soils are comprised of Udic and Ustic Cambosols (Gong et al., 2003), respectively, derived from loamy alluvium. In addition, these three typical areas are characterized by a typical continental monsoon climate with an annual average temperature and precipitation of about 15.4 °C and 1100 mm in Nanjing, 14.5 °C and 831.7 mm in Tongshan and 12.4 °C and 593.8 mm in Shouguang, respectively.

The development of GVP in Nanjing was rapid and formed in geographic clusters around production bases with unified management. Four bases of them, named CB1, CB2, CB3, and OB, were selected for the research. The abbreviation CB meant conventional farming bases where a combination of inorganic and organic fertilizers was used, while OB meant organic farming bases where only organic fertilizers were applied due to the organic-vegetable-oriented production strategy. The durations of greenhouse vegetable cultivation in CB1, CB2, OB, and CB3 were 4, 6, 10 and 12 years, respectively.

Tongshan (CBT) and Shouguang (CBS) are the major vegetable production bases in eastern China as well. Vegetable production in these two study sites typically follows a small family business model using primarily conventional farming methods. With similar farming practices in greenhouse vegetable plots in Tongshan and Shouguang, two study areas could be considered as a big greenhouse vegetable base respectively. The durations of greenhouse vegetable cultivation in CBT and CBS were 30 and 25 years, respectively. Additionally, greenhouse vegetables were primarily cultivated in vegetable–vegetable rotation in the study bases except CBT, where production was conducted in both vegetable–rice and vegetable–vegetable rotations.

In the field survey, the details of types and amounts of fertilizer application were investigated as well. The major inorganic and organic fertilizers used in GVP were composite and manure, of which the application amount varied within 2.9–3.3 and 15–450 t ha^{-1} $year^{-1}$, respectively. Although Cd, Cu, Pb, and Zn contents in the fertilizers were limited with varying from 0.01 to 6.3, 0.6 to 66.9, 0.08 to 27.2 and 2.9 to 438.7 mg kg^{-1}, respectively (Yang et al., 2013), heavy fertilization inevitably introduced considerable amounts of metals to GVP plots in the selected sites.

2.2. Sample collection and treatment

The sampling was performed in the fall of 2011 and spring of 2012. A total of 523 topsoil samples (0–20 cm in Nanjing and Tongshan, 0–30 cm in Shouguang) were collected from both greenhouse plots (441 samples) and open fields near greenhouses (82 samples) using a stainless-steel auger. The difference of sampling depth was due to the difference of root effective depths among the study areas. In Nanjing and Tongshan, cultivated vegetables were mainly leafy vegetables with short root length while fruit vegetables with long root length in Shouguang. A total of 132 greenhouse edible vegetable samples at corresponding soil sampling sites, including fruit, leaf and root vegetables (Table S1) were obtained. Each soil or vegetable sample consisted of five subsamples and was placed in a polyethylene bag. The longitude and latitude of each sampling site was recorded using a hand-hold global positioning system.

All soil samples were air-dried at room temperature. Each soil sample was divided into two parts. One part was ground to pass through a 2.0-mm mesh nylon sieve for the determination of pH and extractable heavy metal contents while another part was ground to pass through a 0.15-mm mesh for organic matter and total heavy metal contents analysis. All fresh vegetable samples were thoroughly washed with tap water, rinsed with de-ionized water and primarily air-dried at room temperature. And then the fresh weight (FW) of the vegetable samples was recorded. After oven dried at 60 °C, the samples were weighed again, and were then ground to powder for further analysis.

2.3. Sample analysis

Soil pH was measured in a paste with a 1:2.5 of soil to water ratio using a glass electrode pH meter (Model PHS-3C, Shanghai Precision and Scientific Instrument Co. Ltd.). Soil organic matter (OM) was analyzed using the Walkley-Black method (Nelson and Sommers, 1982). To determine total Cd, Cu, Pb, and Zn contents, soil was digested with $HClO_4$, HNO_3 and HF. For the determination of Cd, Cu, Pb, and Zn in vegetable, the dried sample was first digested by microwaving with a mixture of HNO_3 and H_2O_2. For the determination of soil available Cd, Cu, Pb, and Zn, the dried soil sample was extracted with 0.05 M $CaCl_2$ (1:2 of soil:extract ratio, 0.5 h