



# Heavy metals in intensive greenhouse vegetable production systems along Yellow Sea of China: Levels, transfer and health risk



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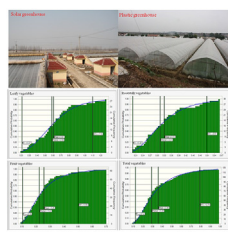
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## HIGHLIGHTS

- Greenhouse vegetable production decreases soil pH, increase soil organic matter and heavy metal contents.
- Leafy vegetables have higher heavy metal transfer factor and health risk compared to other vegetables.
- Rootstalk and fruit vegetables are more suitable for cultivation in greenhouse soils.
- Soil threshold values obtained may help future revision of environmental standards for greenhouse soils.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Recently, greenhouse vegetable production (GVP) has grown rapidly and counts a large proportion of vegetable production in China. In this study, the accumulation, health risk and threshold values of selected heavy metals were evaluated systematically. A total of 120 paired soil and vegetable samples were collected from three typical intensive GVP systems along the Yellow Sea of China. Mean concentrations of Cd, As, Hg, Pb, Cu and Zn in greenhouse soils were 0.21, 7.12, 0.05, 19.81, 24.95 and 94.11 mg kg<sup>-1</sup>, respectively. Compared to rootstalk and fruit vegetables, leafy vegetables had relatively high concentrations and transfer factors of heavy metals. The accumulation of heavy metals in soils was affected by soil pH and soil organic matter. The calculated hazard quotients (HQ) of the heavy metals by vegetable consumption decreased in the order of leafy > rootstalk > fruit vegetables with hazard index (HI) values of 0.61, 0.33 and 0.26, respectively. The HI values were all below 1, which indicates that there is a low risk of greenhouse vegetable consumption. Soil threshold values (STVs) of heavy metals in GVP system were established according to the health risk assessment. The relatively lower transfer factors of rootstalk and fruit vegetables and higher STVs suggest that these types of vegetables are more suitable for cultivation in greenhouse soils. This study will provide an useful reference for controlling heavy metals and developing sustainable GVP.

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## 1. Introduction

Greenhouse vegetable production (GVP) has grown rapidly and

has become a big sector of vegetable production in China due to increasing demand of vegetable consumption and economical benefits for vegetable producers (Hu et al., 2014a; Xu et al., 2015). The intensive management involved in GVP such as fertilization, large cropping index, and high temperature has significantly caused ecological and soil problems such as heavy metals accumulation in greenhouse soils (Hu et al., 2014a; Yang et al., 2014). Accumulation of heavy metals in agricultural soils not only causes soil contamination, but also affects food quality and safety (Jolly et al., 2013; Rafiq et al., 2014). Recently, there has been an increasing concern over heavy metals' accumulation in food and their potential risks to human health (Sun et al., 2013). Thus, it is necessary to systematically evaluate heavy metal uptake by vegetables and assessing the related health risk.

The food chain is one of the most important human exposure pathways to heavy metals (Jan et al., 2010). To ensure safe levels of heavy metals in food special attention has been paid to establish thresholds of heavy metals' concentrations in soil (Sun et al., 2013). The current environmental quality standard applied in China for environmental evaluation and management of GVP does not consider the factors such as specific crop, phytoavailability and relevant human health risk of heavy metals. However, uptake and transfer of heavy metals through vegetables are affected by the concentration of heavy metals in soils, soil properties, vegetable types and species (Rafiq et al., 2014; Xu et al., 2015; Yang et al., 2015). Therefore, setting up a single standard of heavy metals for the large area of GVP in China could over-estimate or underestimate the heavy metals contamination and the associated risk.

To calculate the soil threshold values (STVs) of heavy metals in agricultural system, it is essential to understand the transfer of heavy metals between soil and plant. Some models have been developed to describe the transfer of heavy metals from soil to crop, and for estimating STVs for heavy metals in China (Zhao et al., 2006; Yang et al., 2009). However, those models were developed on hydroponic culture/greenhouse experiments with metal-spiked soils which is different from real agricultural soils, especially GVP soils (Sun et al., 2013; Rafiq et al., 2014). Therefore, it is of importance to develop a relationship between concentrations of heavy metals in soils and their transmission to the edible parts of plants based on the field data to improve the standards of soil environmental quality (Sun et al., 2013). Moreover, in order to assess the risk of agricultural management in GVP system, it is urgently needed to establish the STVs of heavy metals based on soil types and crop absorption characteristics (Hu et al., 2014b).

Half of the World's total vegetable production areas are located in China. By 2013, China's GVP has occupied more than 3.7 million hectare, and become the main type of vegetable production especially along the coast of Yellow Sea with large population demanding for vegetable consumption (CMA, 2015). Previous studies on greenhouse soils in this area have focused on the characteristics of soil organisms, changes of soil properties, nutrition, and salinization (Hu et al., 2014a), and heavy metal accumulations in soils and vegetables (Chen et al., 2013; Yang et al., 2014). However, to our knowledge, there is no comprehensive study explaining the accumulation, transfer and health risk of heavy metals in GVP system from different planting pattern based on a field investigation over regional scale. Furthermore, the STVs of heavy metals in GVP system based on a field survey and health risk assessment have not been reported so far. Therefore, it is imperative to assess the potential risk of heavy metals' accumulation and establish the STVs of heavy metals to provide preliminary data for developing appropriate strategies controlling heavy metals in intensive GVP system.

In order to ensure food safety, it is important to understand the accumulation status, affecting factors, and health risk of heavy

metals in greenhouse soils and vegetables. The aim of this study was to elucidate the accumulation level, transfer characteristic and potential health risk of selected heavy metals from typical intensive GVP systems across the coast of Yellow Sea in China. Further, the STVs of heavy metals were established based on a field survey and health risk assessment. The result of this study will provide a scientific basis to facilitate safety production and sustainable development of GVP in China.

## 2. Materials and methods

### 2.1. Description of the case study areas

In general, greenhouse plots in China are categorized in two types as solar greenhouse (SG) (mainly distributed in the northern parts of warm regions) and plastic greenhouse (PG) (mainly distributed in the southern parts of subtropical regions) (Fig. S1). This study was carried out along the coast of Yellow Sea in China where both SG and PG are prevalent. In this region, greenhouse plots are using intensively to produce vegetables, accounting for 41% of the total GVP area in China (CMA, 2015). Three different GVP bases in Shandong Province, North China, and Jiangsu Province, Southeast China, were selected representing the main types of greenhouse productions along the coast of Yellow Sea in China (Fig. 1).

The first site is located in Shouguang City (SDSG), Shandong Province, with 25 years history of GVP, dominated by SG production systems (Table S1). This city is located in the middle of latitude zone with a warm temperate continental monsoon climate. The soil parent material is composed of alluvium from Mount Tai-Yi, and the soil profiles are classified as Cambosols (CRGCST, 2001; Gong et al., 2003). The second site is in Xuzhou City (JSXZ), Northeast of Jiangsu Province, which is one of the major vegetable production bases in North China with 30 years history of GVP. Both SG and PG vegetable production systems are existed in this area. Topographically, this area is flat and the greenhouse bases are established on Udic Cambosols (CRGCST, 2001; Gong et al., 2003) originated from loamy alluvium. The third site is in Dongtai City (JSDT), Southeast Jiangsu Province and accounting as one of the main vegetable production areas in Southeast China with 20 years history of GVP mainly as PG production system. This site is located in a transition zone between a subtropical temperate zone to a warm temperate monsoon climate zone. The soils are classified as Halosols (CRGCST, 2001; Gong et al., 2003), developed from the marine deposits.

### 2.2. Filed survey and sampling

This study was planned based on a systematic investigation of three GVP systems throughout the above-mentioned areas. The greenhouse plots were chosen according to experience and field survey characteristics such as vegetable types, their planting pattern, planting years, and soil types and properties (Table S1). A total of 120 paired surface soils (0–20 cm) and vegetable samples (104 from greenhouse soils and 16 from open field soils) were collected in summer and autumn 2014. At each point a representative sample was taken composed of five soils/plants' subsamples. Samples were placed in plastic bags and brought to the laboratory for analysis.

Based upon the edible parts, twelve representative species of vegetables were classified into the three following categories: (1) leafy vegetables (Numbers = 28): spinach (*Spinacia oleracea*), cabbage (*Brassica oleracea* var. *capitata*), pakchoi (*Brassica chinensis* L.), and celery (*Apium graveolens* var. *dulce*), (2) rootstalk vegetables (N = 30): radish (*Raphanus sativus* L.), carrot (*Daucus carota* L. var. *sativa* DC.), asparagus lettuce (*Lactuca sativa* var. *angustana* L.), and

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