Effect of greenhouse vegetable farming duration on Zinc accumulation in Northeast China

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Abstract. Greenhouse vegetable production (GVP) has rapidly expanded, and requires more attention due to its heavy metal contamination. In this study, different cultivation greenhouses of 1, 2, 3, 5 and 13 years were selected to investigate the effects of GVP duration on Zn accumulation. The results revealed high Zn (total Zn and available Zn) accumulation in GVP surface layers (0-20 cm), and Zn contents in 0-20 cm soil layers were positively correlated with GVP duration (P<0.01). Zn accumulation was mainly attributed to manure fertilizer application due to higher concentrations of Zn in manures. For greenhouse sustainability, reduction of manure application and reasonable use of passivation materials may alleviate metal phytoavailability and the health risk.

1. Introduction

Greenhouse vegetable production (GVP) has expanded rapidly [1] to meet the increasing market demands of consumers and economic benefits for vegetable producers [2, 3], especially in northeast China where conventional vegetable cultivation is shorter. However, to achieve high vegetable production and economic benefits, some high-intensive managements such as fertilization, irrigation and spraying pesticides have been adopted, which has caused heavy metal accumulation, particularly Cu, Zn, Cd, As and Pb in greenhouse vegetable production soils (GVPS). Accumulation of heavy metals in GVPS not only causes soil contamination but also elevates metal phytoavailability, resulting in potential risk to human health. Recently, there has been an increasing concern over the security associated with greenhouse environment safety due to the increasing vegetable consumption. Several researches have assessed heavy metal uptake by greenhouse vegetables and subsequent health risk [4, 5], but studies on health risk changes with GVP duration were less conducted. Soil total and available metals are important to predict metal phytoavailability and toxicity [6, 7]. Moreover, cultivation years of greenhouses may cause metal accumulation in soils. Therefore, the objects of this research were to investigate the distribution characteristics of Zn in GVPS and illustrate the effects of GVP duration on metal accumulation.

2. Materials and Methods

2.1. Area of investigation
The survey region is an important vegetable planting areas in Northeast China, located at 41°55'N and 122°58'E. It occupies 113.7 km², situated in Shenyang suburb. There are numerous agricultural facilities in this study area. About 100 t·hm⁻² manures (e.g. decaying chicken and dairy manures), 1000 kg·hm⁻² potassium sulphate (K₂SO₄), 1000-1500 kg·hm⁻² diammonium phosphate ((NH₄)₂HPO₄), 500 kg·hm⁻² carbamide or 1000 kg·hm⁻² ammonium sulphate ((NH₄)₂SO₄) were utilized each year.

2.2. Sampling and analysis
This study was carried out in August at 41 sample sites in different cultivation greenhouses of 1, 2, 3, 5 and 13 years. Soil samples were also collected from the open vegetable field (OF), serving as the control (CK). The depth of soil sample was 0-120 cm. Each sample was multi-point mixed.

Soil samples were air dried at room temperature, passed through 2-mm nylon sieves, mixed carefully and stored for Zn analysis. Prior to total Zn analysis, the samples were ground to powders, sieved using 0.25-mm nylon sieve. About 0.2000 g of sample was digested in an HCl-HNO₃-HClO₄ (3:1:1, v/v) mixture, and determined by using the atomic absorption spectrophotometer (WFX-120, Beijing Rayleigh Analytical Instrument Corp, China). Plant available Zn was determined by using diethylene-triamine-pentacetic (DTPA) buffered at pH 7.3[8].

Statistical analysis was performed using SPSS 19.0. One-way ANOVA was used to analyze the effects of cultivation years on Pb accumulation in greenhouses soils using the least significance at 5% level.

3. Results

3.1. Distributions of available Zn in soil profiles
DTPA-Zn is important to predict metal phyto-availability, and can be identified as available and potential toxic to the environment. DTPA-Zn concentrations in soils are shown in Figure 1. DTPA-Zn concentrations decreased with soil depth in the order of 0-20 > 20-40 >40-120. It was found that DTPA-Zn accumulated in 0-40 cm soils, and increased with utilization years of GVP. According to the results of analysis of variance of DTPA-Zn in GVP surface layer (0-20cm) and OF surface layer (0-20cm), there were significant differences (P<0.05). The mean concentrations of DTPA-Zn in GVPs were higher than those in the open vegetable field soils (OFS). The contents of plant available Zn peaked in 0-20 cm soil profiles of 13-year GVP, and reached 9.24±1.62 mg·kg⁻¹. In 0-20 cm soils,
DTPA-Zn contents in OFS were significantly lower than those in GVPS (except 1-year GVP) (P<0.05). In 20–40 cm soil profiles, DTPA-Zn contents of 13-year and 5-year GVP were still significantly higher than those of OF (P<0.05).

As shown in Figure 2, total Zn in soils decreased with soil depth, and accumulated in 0–40 cm soil profiles. The mean contents of GVPS were higher than those in OFS. Total Zn in GVPS showed an increasing trend with growing GVP duration in the research region, especially in surface soils (0–20 cm), however, there was no similar trend in 40–120 cm soil profiles. As for 0–20 cm soil profiles, there was no significant differences among 2-year, 3-year and 5-year GVPS, but total Zn concentrations in 2-year, 3-year and 5-year GVPS were significantly higher than those in OFS (P<0.05). The maximum contents of total Zn were found in 0–20 cm soil profiles of 13-year GVP, and the value was 59.80±11.37 mg·kg⁻¹.

Available Zn (DTPA-Zn) and total Zn in GVP surface layer (0–20 cm) showed a similar relationship with cultivation duration (Figure 3). As for available Zn, DTPA-Zn contents increased with GVP

Figure 3. Temporal trends of DTPA-Zn and total Zn in GVP surface layer (0–20 cm). The error bars indicate the standard deviations.
duration, and positively correlated with cultivation years ($R^2=0.828$, $P<0.01$). As for total Zn, total Zn concentrations also increased with the planting years of GVP, and there was a positive correlation between Zn contents in soils and cultivation duration ($R^2=0.910$, $P<0.01$). The variability of available Zn or total Zn in GVP utilized for more than 5 years was larger than those in greenhouses cultivated for less than 4 years. The results were corresponded with Lanqin Y[9].

4. Discussion
Based on increase of Zn contents (including available Zn and total Zn) in surface layers (0-20 cm) with growing GVP cultivation (Figure 3), it can be concluded that increasing GVP duration could cause metal accumulation. Cultivation years also affected total Zn contents in deeper soil layers (40-120 cm), mostly due to redistribution of trace metals in soils under water movement, biological activities and geochemical reactions [10]. The concentrations of available Zn and total Zn in GVPS were both higher than those in OFS, especially in surface layers (Figure 1, Figure 2). This suggested that greenhouse soils had larger enrichment ability for Zn, which was in accordance with the results of Lan et al. [11] and Wen et al. [12]. Trace metals were likely to have primarily originated from anthropogenic sources (e.g. fertilizer application, atmosphere deposition, wastewater irrigation, traffic emission et al.), other than the pedogenic weathering of primary minerals. In our research region, there was no industrial pollution source, and the research greenhouses were far away from motor traffic. So Zn in greenhouse soils can be identified as arising from agricultural activities, such as manure and fertilizer application. The owners of greenhouses consisting of local farmers applied excessive manures and fertilizers in order to elevate production. According to our comprehensive investigation, the rate of manure fertilizer ranged from $100\times10^3$ kg·ha$^{-1}$ to $120\times10^3$ kg·ha$^{-1}$ each year. Zn contents of manures ranged from 408 mg·kg$^{-1}$ to 889 mg·kg$^{-1}$. Based on these results of investigation, it could be calculated that manure fertilizers can contribute $40.8-106.7$ kg·ha$^{-1}$·year$^{-1}$. However, Zn contents of chemical fertilizers were much less than those of manure fertilizers, and they had been neglected. Therefore the long term use of manure fertilizers can cause Zn accumulation in greenhouse soils. It can be explained that duration of land use as greenhouse and management practices are are more important factors [13, 14].

5. Conclusion
In northeast China, cultivation years have caused Zn accumulation in GVPS, especially in surface layers (0-20 cm). Available Zn may pose a potential risk to human health, which may result in metal uptake by greenhouse vegetables. The application of manure fertilizers can cause much greater increase of Zn in GVPS. For greenhouse sustainability, reduction of manure application and reasonable use of passivation materials may alleviate metal phytoavailability and the health risk.

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