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# Accumulation Characteristics and Sources of Available Pb in Soils under Greenhouse Cultivation Condition in Northeast China

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**Abstract.** Greenhouse vegetable production (GVP) is one of the most effective cultivation methods, and the enrichment of heavy metals has been found in greenhouse soils. This study was conducted to determine the influence of cultivation ages on available Pb and find out pollutant sources. The concentrations of available Pb decreased with soil depth, and obviously accumulated in 0-40 cm soil profiles ( $P < 0.05$ ). Available Pb contents in 0-40 cm soil profiles fluctuated and did not significantly ( $P > 0.05$ ) correlate with cultivation duration. According to correlation analysis, there was no significant correlation ( $P > 0.05$ ) between available Pb and soil properties (pH, SOM, sand, silt, clay,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ ). It can be concluded that Pb in greenhouse soils might arise from comprehensive sources including anthropogenic activities and natural effects. Therefore, agricultural practices (i.e. manure and fertilizer input, pesticide spray, and application of agricultural machines) will not be a dominant factor of available Pb accumulation.

## 1. Introduction

Greenhouse vegetable production (GVP), which is 10 times higher more than the open field farming, has increased rapidly to keep up with growth in population. The area under greenhouses in China is about 85% of the world's greenhouse area<sup>[1]</sup>. The type of intensive agriculture system requires considerable input of labour, fertilizers, pesticides and energy to maximize vegetable yields. However, the intensive application of agricultural chemicals and different agricultural practices (i.e. plow and tillage) has promoted the continuous accumulation of heavy metals in greenhouse soils<sup>[2, 3]</sup>. Soil pollution in greenhouses may elevate metal phytoavailability. Available metals in soils, which are considered to be easily adsorbed by vegetables and accumulated in their edible parts, can pose a greater risk to human health through the food chain than heavily polluted areas<sup>[4]</sup>. Thus, to protect human health and diet safety, it is necessary to understand the accumulation status of available metals and find out the pollutant sources under greenhouse condition.

Lead is a nonessential element for human. And oral ingestion of vegetables polluted by Pb contaminated soil is a major pathway for exposure to humans<sup>[5]</sup>. Once Pb enters to human body, it will be hardly excreted out. Even at a low level in blood, Pb can cause a range of neurocognitive, behavioural and other specific issues<sup>[6]</sup>. Although total Pb concentrations in soils partly contribute to vegetable absorption, available Pb has a stronger connection with Pb contents in vegetable edible parts. Thus, it is necessary to investigate available Pb in soils.



Accordingly, the objects of this study were to (i) explore the migration ability of available Pb in the soil profiles; (ii) reveal the accumulation characteristics of available Pb with cultivation duration; (iii) to find out the potential sources of Pb.

## 2. Materials and Methods

### 2.1. Study area

The study area, located in the west of Shenyang City, in Northeast China, covers a total areas of 113.7 km<sup>2</sup> [7] and produces about 40 billion tons of various vegetables (e.g. cucumber, tomato, and pakchoi) every year [8]. Greenhouses are dominant agricultural facilities for vegetable production in research region. Approximately 80 t·hm<sup>-2</sup> of decaying chicken manure, 500 kg·hm<sup>-2</sup> of carbamide or 1000 kg·hm<sup>-2</sup> of ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>), 1000 ~ 1500 kg·hm<sup>-2</sup> of diammonium phosphate ((NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub>) are used each year. There are no mining companies and manufacture factories in the study area. Only a national highway traverses through it from east to west. The dominant soil is brown soil with pH 7.14±0.46, TOC 8.16±2.45 g·kg<sup>-1</sup>, CEC 12.84±0.94 mg·kg<sup>-1</sup>, the soil bulk density 1.33±0.09 g·cm<sup>-3</sup>.

### 2.2. Sampling and analysis

Greenhouses of 1, 2, 5, 6, 10 and 13 years of consistent cultivation and management were selected, and the adjacent open field served as the control. Soil samples (0-20 cm, 20-40 cm, 40-60 cm, 80-100 cm, 100-120 cm) were randomly collected. Each sample was mixed by 3-5 bore of cores. Soil samples were firstly air dried in laboratory, and then grounded to pass through 0.25-mm and 0.149 nylon sieves for determination of available Pb and soil properties (pH, SOM, sand, silt, clay, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub>).

Available Pb was evaluated by using diethylene triamine pentacetic acid (DTPA) buffered at pH 7.3 [9]. pH was determined by using a combination glass electrode and a conductivity meter in a 1:1 ratio (w/v) of soil: deionised water suspension [10]. Total organic carbon (TOC) was determined according to the dichromate, wet oxidation method described by Walkley and Black [11]. Salt content in soils was determined by gravimetric method. Particle size distribution (texture) of the soil was determined by using air-dried samples according to the pipette method [12, 13].

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

## 3. Results

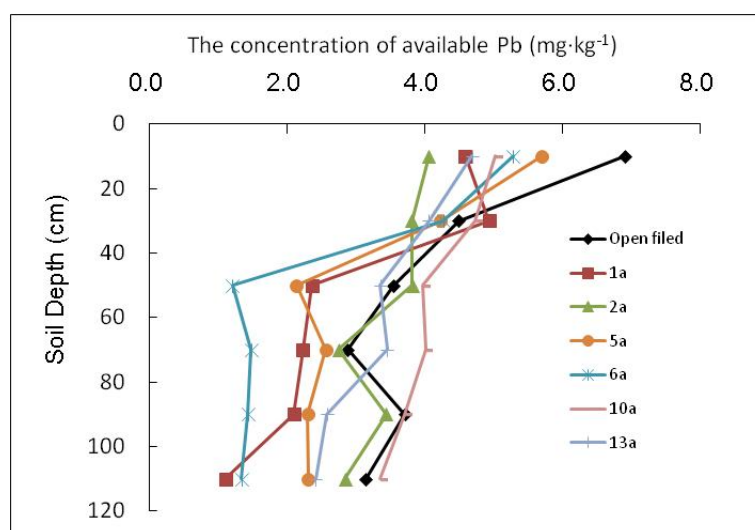


Figure 1. Distribution of available Pb in soil profiles with different planting years

### 3.1. Characteristics of available Pb distribution in soil profiles

Available Pb contents in soils decreased with soil depth, and obviously accumulated in 0-40 cm soil profiles ( $P < 0.05$ ) (Figure 1). According to the two-factor analysis results, soil depth had a significant effect on available Pb concentrations ( $F = 8.962$ ,  $P < 0.01$ ), but there was no prominent correlation between available Pb contents and GVP duration ( $F = 1.078$ ,  $P = 0.382$ ). In 60-80 cm soil profiles, the order of available Pb contents accumulation levels in soils was ranked as 10-a > 5-a > 13-a > 1-a > 6-a > the open field > 2-a. However, available Pb concentrations in 1-year, 13-year, 6-year and 5-year greenhouses (except 10-year greenhouse) were lower than those of the open field and 2-a GVP in 60-80 cm soil profiles. And there was a similar trend of available Pb in soils of 5-year, 6-year, 10-year and 13-year GVP.

### 3.2. Effects of Greenhouse cultivation on available Pb contents

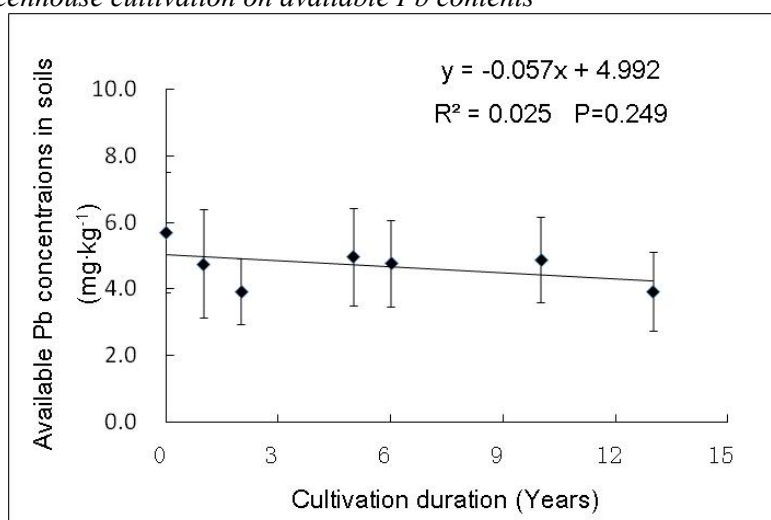


Figure 2. Temporal trends of available Pb in GVP surface layer (0-40 cm).  
The error bars indicate the standard deviations.

Figure 3 showed the relationship of available Pb accumulation with utilization ages of vegetable greenhouses in 0-40 cm layers. There was no significant correlation between available Pb and duration interaction was not significant ( $F = 1.096$ ,  $P = 0.378$ ), implying that the shift in land use did not obviously affect Pb activities. The concentrations of available Pb in the open field were higher than those in the greenhouse soils. There was a decline trend of available Pb with cultivation years, but significantly negative correlations did not exist ( $P = 0.254$ ). The CVs for available Pb in the open field and 1-year greenhouse were relatively higher, with CVs of 31.57% and 34.24% respectively, which may related to different agricultural practices<sup>[14]</sup>.

### 3.3. Correlation analysis

A statistical correlation analysis between available Pb and soil properties (pH, SOM, sand, silt, clay,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ ) of 54 greenhouse soils was performed, and the results are provided in Table 1. Soil organic matters (SOM) were negatively correlated with sand and  $\text{SiO}_2$ , and was positively correlated with silt, clay,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  ( $\alpha = 0.01$  and  $\alpha = 0.05$ ). Positive correlation suggests SOM, silt, clay,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  have common sources, mutual dependence and identical behavior during transport<sup>[15]</sup>. Negative correlation suggests SOM,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$  have various traits, and derive from different sources. However, no significant correlation was observed among available Pb, pH, SOM, sand, silt, clay,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ , suggesting that a link with available Pb and common geochemical characteristics was not close.

Table 1 Correlation coefficients among available Pb and soil properties in greenhouse soils.

	A-Pb	pH	SOM	sand	silt	clay	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>
A-Pb	1								
pH	0.061	1							
SOM	-0.06	-0.047	1						
Sand	0.003	0.072	-0.492**	1					
Silt	0.002	-0.215	0.488**	-0.944**	1				
clay	-0.009	0.142	0.405**	-0.889**	0.689**	1			
SiO <sub>2</sub>	0.255	-0.09	-0.305*	-0.041	-0.007	0.101	1		
Al <sub>2</sub> O <sub>3</sub>	0.104	0.257	0.335*	-0.513**	0.372*	0.613**	-0.277	1	
Fe <sub>2</sub> O <sub>3</sub>	0.099	0.255	0.335*	-0.516**	0.373*	0.618**	-0.273	0.873**	1

A-Pb: available Pb contents

Significant levels: \*  $\alpha=0.05$ , and \*\*  $\alpha=0.01$ ; N=54

#### 4. Discussion

The trace metals in agricultural ecosystem primarily originated from human inputs through manure application, pesticide spray and fertilizer utilization [16, 17, 18]. Lead accumulation in soils may be ascribed to pedogenic weathering, automobile exhaust, and industrial manufacture. The greenhouses in our research region are far away from highway, and there was no factory nearby. So automobile exhaust and industrial manufacture have less effect on available Pb accumulation. According to our investigation, 80-100 t·hm<sup>-1</sup> year<sup>-1</sup> chicken and dairy manures were applied to greenhouse vegetable production (GVP). Manures contained more than 40% of organic matter, and organic matters from topsoil were significantly and positively related to manure input. Long-term manure application could significantly ( $P<0.05$ ) increase soil organic C (SOC). It can be concluded soil organic matters (SOM) may accumulate with GVP duration. Positive correlation among SOM, silt, clay, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> can be identified as arising from anthropogenic sources. The manures were more responsible for SOM accumulation. Negative correlation among SOM, sand and SiO<sub>2</sub> can be identified as arising from natural effects, such as pedogenic progress. Lead showed no correlation with soil properties, suggesting that Pb can be identified as arising from anthropogenic and natural sources. Pb in greenhouse soils may derive from ploughing and tillage by agricultural machine and from atmospheric deposition during the summer when plastic mulch is removed. Furthermore, there was no significant relationship between available Pb and duration interaction ( $F=1.096$ ,  $P=0.378$ ), suggesting that human activities did not primarily affect available Pb enrichment. Available Pb accumulation in greenhouse soils can be ascribed to comprehensive sources. Although natural effects can explain part of available Pb accumulation in greenhouse soils, the shift in land use and agricultural activities can partly affect available Pb contents.

#### 5. Conclusion

Available Pb in greenhouse soils decreased with soil depth (Figure 1), fluctuated due to different agricultural activities (Figure 2), and did not significantly correlate with soil properties (pH, SOM, sand, silt, clay, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub>) (Table 1). Soil organic matters (SOM) were negatively correlated with sand and SiO<sub>2</sub>, and were positively correlated with silt, clay, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> ( $\alpha=0.01$  and  $\alpha=0.05$ ) (Table 1). Available Pb accumulation in greenhouse soil can arise from comprehensive sources including anthropogenic sources and natural effects (i.e. pedogenic weathering). Therefore, agricultural practices (i.e. manure and fertilizer input, pesticide spray, and application of agricultural machines) will not be a dominant factor of available Pb accumulation, but their impacts on Pb require further testing in long-term field experiments.

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