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Accumulation, sources and health risks of trace metals in elevated geochemical background soils used for greenhouse vegetable production in southwestern China

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ABSTRACT

Greenhouse vegetable cultivation with substantive manure and fertilizer input on soils with an elevated geochemical background can accumulate trace metals in soils and plants leading to human health risks. Studies on trace metal accumulation over a land use shift duration in an elevated geochemical background scenario are lacking. Accumulation characteristics of seven trace metals in greenhouse soil and edible plants were evaluated along with an assessment of the health risk to the consumers. A total of 118 greenhouse surface soils (0–20 cm) and 30 vegetables were collected from Kunming City, Yunnan Province, southwestern China, and analyzed for total Cd, Pb, Cu, Zn, As, Hg, and Cr content by ICP-MS and AFS. The trace metals were ordered Cu > Cd > Hg > Zn > Pb > As > Cr in greenhouse soils accumulation level, and the geo-accumulation index suggested the soil more severely polluted with Cd, Cu, Hg and Zn. The greenhouse and open-field soils had significant difference in Cd, Cr and Zn. The duration of shift from paddy to greenhouse land-use significantly influenced trace metal accumulation with a dramatic change during five to ten year greenhouse land-use, and continuous increase of Cd and Hg. A spatial pattern from north to south for Cd and Hg and a zonal pattern for Cu and Zn were found. An anthropogenic source primarily caused trace metal accumulation, where the principal component analysis/multiple linear regression indicated a contribution 61.2%. While the assessment showed no potential risk for children and adults, the hazard health risks index was greater than one for adolescents. The extended duration of land use as greenhouses caused the trace metal accumulation, rotation in land use should be promoted to reduce the health risks.

1. Introduction

Agricultural pollution is of increasing concern because of food safety issues and the potential human health risk (Dziubanek et al., 2015; Lu et al., 2015; Mahar et al., 2016). Trace metal accumulation, toxicity and persistence in agricultural soil is a widely studied topic in soil and environmental sciences (Aelion et al., 2008; Rahman et al., 2014). Arsenic, cadmium, lead and mercury can be harmful even at low concentrations when ingested over an extended period of time, triggering numerous illnesses including cardiovascular disease and cancer (Bahemuka and Mubofu, 1999; Järup, 2003; Sharma et al., 2009; WHO, 1995). The trace metals released by weathering of parent rock and pedogenesis are redistributed in the soil under water movement, biological activities and geochemical reactions (Rodríguez et al., 2013). Over the last few decades, anthropogenic activities including agricultural practices, mining and smelting and atmospheric deposition have caused the trace metal pollution in soil (Kelepertzis, 2014; Li et al., 2011; Pourrut et al., 2011; Wu et al., 2012). Several trace metals in agricultural soils from human input have exceeded the natural input in some areas (Facchinelli et al., 2001).

Greenhouse vegetable production in China increased rapidly to meet the growing demand for vegetables. The area under greenhouses in 2010 was 4.7 million ha, about 85% of the world's greenhouse area (Yang et al., 2014a). The greenhouse is a labor- and energy-intensive production system (Holvoet et al., 2015), fertilizers and pesticides are applied excessively to maximize vegetable yields and economic bene-

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fits. High-intensity cropping and management practices cause trace metal accumulation in greenhouse vegetable soils, and the trace metal enrichment in vegetables. The trace metals in greenhouse soils of China have been studied (Chen et al., 2014; Liu et al., 2011, 2015; Qi et al., 2005).

While there is a rapid increase in area under greenhouse vegetable production (Moritsuka et al., 2013; Zhang et al., 2013) where leafy vegetables are prevalent, the region of Kunming city is known for nonferrous metal resources where reserves of zinc, copper, lead and phosphate ore are high (Yang et al., 2012; Wang et al., 1983, 2015). The source rocks from which parent material in Yunnan Province is derived contain high trace metal concentrations. The geochemical background concentrations of As, Cu, Cd, Hg and Pb are 2.2, 2.1, 1.9, 1.7 and 1.6 times higher, respectively, than the respective national average value (Terminal Station of Environmental Monitoring of China, 1990). Therefore, the anthropogenic and natural sources can jointly induce trace metal accumulation in the greenhouse soils, potentially increasing concentration in the food chain. The soil trace metal accumulation in greenhouse soils with high geochemical background values has been less well documented in previous studies. Thus, the study of soil trace metal accumulation in greenhouse soils from Kunming City is of real-world relevance.

The objectives of this research were to (1) investigate trace metal concentrations and accumulation in the greenhouse soils of Kunming City, Yunnan Province, southwestern China; (2) determine the lithogenic and anthropogenic contributions of trace metal accumulation in the study area; and (3) assess the potential health risk to inhabitants through vegetable consumption in areas with elevated geochemical background and intensive cultivation.

2. Materials and methods

2.1. Study area

The Guandu-Chenggong-Jinning belt is located on the east bank of Dianchi Lake in Kunming City, Yunnan Province, southwestern China. The elevation varies from 1880 m to 1986 m above the sea level from the south to the north (Fig. 1). A subtropical humid monsoon climate prevails in the area with a mean annual temperature of 15 °C and precipitation 1035 mm, of which 90% falls between May and October. Ferric Acrisol (FAO, 1998) derived from the limestone, sandstone residuum, slope wash and alluvium are the dominant soils in the area. The crop production in the study area shifted from rice cultivation to greenhouse vegetable production during the past 30 years.

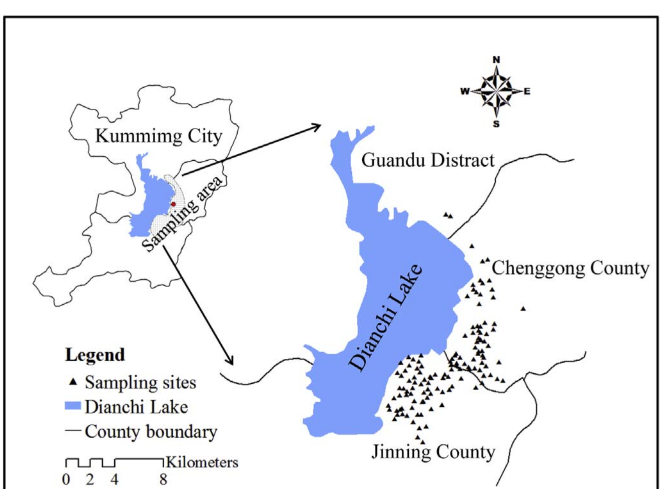

Fig. 1. Location of sampling sites in Kunming City, Yunnan Province, Southwestern China.

2.2. Sample collection and field investigation

Soil and plant samples were collected from 118 greenhouses and 11 open vegetable fields during December 2014. A stainless-steel auger was driven to 20 cm depth at five locations in a greenhouse to make a composite sample. Thirty edible vegetables including tomato, eggplant, broccoli, celery, spinach, swamp cabbage, Chinese cabbage, lettuce, and pakchoi were sampled at the corresponding soil sampling sites. To determine the soil geochemical background for the study area, twenty randomly selected soils were sampled at 80–100 cm depths. In addition, six organic fertilizers including cow manure, pig manure, sheep manure and chicken manure were sampled. A hand-held global positioning system receiver was used to record the position of the sampling sites.

Information on the vegetable rotation pattern, fertilization, irrigation, vegetation species, pesticides applied and historical data were recorded from the vegetable growers using a structured questionnaire. Temporal images were obtained for the 1990–2014 from Google Earth satellite to determine the land use change against the questionnaire responses.

2.3. Sample treatment and analysis

Soil and manure samples were air dried, and stones and plant debris were handpicked and ground to pass through a 2 mm nylon sieve. A part of ground sample was passed through a 0.149 mm nylon sieve for As, Cd, Cr, Cu, Hg, Pb and Zn analyses. Plant was washed with tap water followed by deionized water to remove airborne pollutants, and fresh weight was recorded. Plant surface water was dried at 65 °C until a constant weight, and ground into powder for trace metals analysis.

The soil pH was measured in a 1:2.5 soil-water suspension (Lu, 1999). The soils were digested in HNO_3-$HClO_4$-HF (Zhuang et al., 2009), and analyzed for Cd, Cr, Pb, Cu and Zn using inductively coupled plasma mass spectroscopy (ICP-MS; American Thermo Scientific, X7). For As and Hg measurement, the soil was digested in aqua regia (HNO_3:HCl = 3:1) and analyzed by atomic fluorescence spectrometry (AFS; Beijing Jitian Instruments Co., Ltd. production, AFS-820) (Xu et al., 2013). Plant and organic fertilizer were digested in HNO_3-$HClO_4$ (5:1) and the trace metals were quantified via AFS and ICP-MS (Allen et al., 1986).

2.4. Parameter estimation

Geo-accumulation index (I_{geo}), vegetation enrichment factor (VEF), target hazard quotients (THQs) and hazard index (HI) were calculated from the data to evaluate the accumulation of the trace metals and health risks. The geo-accumulation index, a geochemical criterion for evaluating the pollution level (Muller, 1969):

$$I_{geo} = Log_2\left(\frac{C_n}{1.5 \times B_n}\right) \quad (1)$$

where C_n is the measured concentration of trace metal, B_n the geochemical background value of the trace metal, the mean concentrations at the 80–100 cm depth. Coefficient 1.5 of possible variations in background associated with lithological variations (Stoffers et al., 1986). A site was classed as unpolluted with $I_{geo} \le 0$; unpolluted to moderately polluted with $0 < I_{geo} \le 1$; moderately polluted with $1 < I_{geo} \le 2$; moderately to heavily polluted with $2 < I_{geo} \le 3$; heavily polluted with $3 < I_{geo} \le 4$; heavily to extremely polluted with $4 < I_{geo} \le 5$; and extremely polluted with $I_{geo} > 5$ (Chen et al., 2014). The vegetable enrichment factor, defined as the ratio of the trace metal concentration in vegetables to that of the soil (Khan et al., 2010):

$$VEF = C_{veg}/C_{soil} \quad (2)$$

where C_{veg} and C_{soil} represent the trace metal concentration in vegetables (fresh weight) and soils (dry weight), respectively. The