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Bioaccumulation and sources of metal(loid)s in lilies and their potential health risks

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A B S T R A C T

Dietary intake of metal(loid)s can seriously affect human health, but the levels, the bioaccumulation, sources and related health risks of As, Cd, Cr and Pb in cultivated lilies, particularly for *Lilium davidii* var. *unicolor*, remain unresolved. We collected 35 lily samples aged 1–6 years from farmlands of two types of soil (heilu soils and loessal soils) in Qilihe district in 2016 and analysed the concentrations of As, Cd, Cr and Pb in bulbs, the soil-bulb bioaccumulation and the potential sources of these elements in bulbs. Non-carcinogenic and carcinogenic risks by consuming lilies were also assessed. Concentrations of four elements decreased in the order of Cr > Pb > Cd > As, and soil-bulb BCFs in the order of $BCF_{Cd} > BCF_{Cr} > BCF_{Pb} > BCF_{As}$. The Cd concentration of bulbs of lilies which grew in heilu soils was statistically higher than that of bulbs of lilies which grew in loessal soils, and the Cd concentration of bulbs of lilies aged 1–3 years was statistically higher than that of bulbs of lilies aged 4–6 years. Levels and soil-bulb BCFs of Cr and Pb of two-bulbed lilies were statistically higher than those of one-bulbed lilies. Farmyard manure may be a primary source of Cd in soil. There existed overall potential non-carcinogenic effects by exposure to the combination of four elements. Dietary intake of Cr posed carcinogenic risks to both adults and children. Non-carcinogenic and carcinogenic risks were higher for adults than children. Concluding, the edible parts of lily were significantly polluted by Cr and Pb but not by As and Cd. The number of bulbs significantly impacted concentrations and soil-bulb BCFs of Cr and Pb, but the reason for which needs further studies. Non-carcinogenic and carcinogenic risks caused by lily consumption should not be neglected.

1. Introduction

Food safety has received increasing attention with the rapid development of the economy. Dietary intake of metal(loid)s, which are introduced to food products by agricultural production and other human activities, can seriously affect human health. The bioaccumulation of metal(loid)s causes carcinogenic, mutagenic, and teratogenic effects. Chronic Pb exposure is toxic to every human organ system, especially the renal, reproductive, and nervous systems, and children are particularly sensitive to Pb exposure and can suffer neurological problems when exposed to considerably lower Pb levels than adults (Meyer et al., 2008; Zhang et al., 2012). Cd is not easily excreted from the body and therefore accumulates over one's lifetime, causing kidney damage and skeletal abnormalities (Lauwerys et al., 1994; Satarug et al., 2003). Dietary intake of Cr may cause breast cancer (Pasha et al., 2010). In addition, Exposure to Cr (III) can induce the oxidative stress and genotoxicity, and may cause some health problems, such as respiratory illness, diabetes, gastrointestinal tract problems and dermal problems, etc (Khan et al., 2012). Chronic exposure to As, especially inorganic As (henceforth denoted iAs), leads to skin, bladder, and lung cancers (Carlin et al., 2016; James et al., 2015; Naujokas et al., 2013). Factors that influence the concentrations of metal(loid)s in plants include environmental pollution, nature of the soil in which the plant is located, and bioaccumulation ability of the plant (Seyfferth et al., 2016; Wang et al., 2006). Fertilizers that contain heavy metals can pollute soil, thereby becoming an important potential source of metal(loid)s in plants (Xu and Zhang, 2017).

Lilium davidii var. *unicolor* (primarily produced in Lanzhou), a perennial herb, is a nutritious vegetable with high dietary and medicinal values. *L. davidii* var. *unicolor*, which is fleshy and delicate, is high in sugar and low in crude fibre and contains many other beneficial ingredients (Li et al., 2012; Li et al., 2007; Li, 2007). Mature lilies are usually consumed after cooking, such as stir-frying and cooking porridge. The growth cycle of *L. davidii* var. *unicolor* is relatively long. It usually takes about six years to mature from the small bulbs (commonly known as "mother seeds"). It is suitable to use rotation cultivation for *L. davidii* var. *unicolor* (Wang et al., 2011). The levels of nutrients, such as Mo, Zn, Cu, CaO, B and Na_2O, in the farmlands in which 1-year-old, 2-

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year-old and 3-year-old lilies grow are lower than those in the unplanted farmlands. In addition, the levels of N, P, Zn, Cu, Pb, Se, MgO, B, I and the organic matter in the farmlands in which 3-year-old lilies grow are lower than those in the farmlands in which 1-year-old and 2-year-old lilies grow. Therefore, due to the degradation of soil quality, lilies should be transplanted to another farmland after they have grown for 3 years in one farmland (Kuang et al., 2010), and it would be better that the original farmland is used to plant grass and leguminous crops (Zhou, 1986). Usually, the "mother seeds" are harvested while the mature lilies are harvested. Small "mother seeds" weighing about 4 g are bred in the "mother seed" farmland, and all of them are dug out after 2–3 years. Then, bulbs of one-bulbed lilies, which are more than 15 g, are selected and transplanted from the "mother seed" farmland to another farmland. After about 3 years, they grow up to more than 150 g, which are mature lilies, and are harvested. Before *L. davidii* var. *unicolor* is planted, fully decomposed fertilizers (livestock manure and human waste) are spread all over the farmland by 45,000–60,000 kg hm^{-2} according to the soil fertility conditions, and then the farmland is deeply turned over to make the fertilizers even. Moreover, during the growth of the plant, topdressing (livestock manure and human waste) is deeply applied between the rows of lilies by 15,000 kg hm^{-2} when the height of the plant reaches about 20 cm, to promote the growth of the bulbs (Wei, 2017; Hou, 2016; Wang et al., 2011). There have been notably few studies investigating metal(loid)s in *L. davidii* var. *unicolor*. Li et al. (2015) analysed the potential ecological and health risks of Cd and Pb in *L. davidii* var. *unicolor* and in the soil in which the lilies grow in Gansu province using the potential ecological risk index and target hazard quotient. In addition, Yang et al. (2014) reported the concentrations of Pb and Cd in three drying products and three dried products. However, existing studies primarily examine Cd and Pb, with relatively little attention being given to As and Cr. The sources of metal (loid)s in the bulb and the controls on the concentrations of metal(loid)s in *L. davidii* var. *unicolor*, such as age of lily and soil type, have also not been considered in these studies. Moreover, there is little research that investigates the accumulation of metal(loid)s from the host soil to the edible part of lily and the health risks associated with the ingestion of lily.

Qilihe District, located in the south part of Lanzhou, is at an elevation of approximately 1800–2200 m. The climate is cool and humid with extensive vegetation coverage. The annual average temperature is 5.8 °C and the accumulated temperature that is not less than 10 °C is 1128–2349 °C. The average frost-free period of the year is 135 days. The average annual rainfall is 460 mm. The growth of *L. davidii* var. *unicolor* requires a unique ecological environment, including the proper elevation, temperature, precipitation, soil, and slope (Chen et al., 2003). Due to the climate of Qilihe District, *L. davidii* var. *unicolor* is one of the most characteristic agricultural products in Lanzhou and is a primary industry in mountainous areas that have formidable natural conditions. In recent years, the cultivated area of lily in Qilihe District has reached 3640 hm^2, increasing by 36.5% from 2012. The annual production of lily has reached 2.82 million tons, which is 88% more than that in 2012. In 2016, the export volume of lily reached 300 t (She, 2012; Zhou, 2016; Qilihefabu, 2017).

The main objectives of this study are to investigate the levels of As, Cd, Cr and Pb in *L. davidii* var. *unicolor*, and analyse the transfer ability of metal(loid)s from soil to the edible part of lily, sources of metal(loid) s in bulbs and related health risks of metal(loid)s.

2. Methods and materials

2.1. Sample collection

Samples were collected between the end of August and early September 2016. The farmlands of different soil types (heilu soils and loessal soils) were randomly selected in Qilihe District as the sampling points. The lilies of different ages were separately planted in different farmlands. Thirty-five lily samples, 1–6 years old, were collected from these farmlands with a stainless steel shovel respectively. The detailed information about the samples were shown in Table S1. Organic fertilizers (chicken manure, cow manure and sheep manure) were also collected. Samples were stored in polyethylene bags and were transported to the laboratory.

2.2. Sample preparation and analysis

Rhizospheric soil was shaken from the plant and collected for analysis. The bulbs were separated from plants and washed three times with tap water and three times thereafter with ultrapure water. The washed bulb samples were freeze-dried, ground and passed through 100 mesh nylon sieve. Soil samples were also freeze-dried, ground and passed through 20 mesh nylon sieve. Each soil sample was divided into two portions using the quartile method. One sample was further ground and filtered through 100 mesh nylon sieve for the analysis of metal (loid) concentrations, and the other sample was used for the measurement of soil pH.

Soil samples and ultrapure water were added to beakers at a mass ratio of 1:2. The mixture was stirred well with a glass rod. After standing for 30 min, soil pH was determined using a pH 400 m (Spectrum, USA).

After pretreatment, 0.1 g of soil sample was digested in a poly-tetrafluoroethylene jar with a mixture of HNO_3 (5 mL)-HF (2 mL)-$HClO_4$ (1 mL). The mixture was heated to 120 °C for 1 h, heated to 140 °C for 1 h, heated to 160 °C for 1 h, and heated to 180 °C for 45 min in an automatic digestion instrument (ST-60, Beijing Pu Tyco Instrument Co., Ltd). The reflux funnel was later removed, and the acid was dried. The digested liquid was cooled to room temperature and subsequently diluted with ultrapure water to 25 mL. Next, 0.2 g of lily sample, which had been sieved through 100 mesh nylon sieve, was placed in a Teflon jar, where 2 mL HNO_3 (MOS grade) was added. After cold digestion for 4 h, the Teflon jar was placed into a stainless steel sleeve and tightened, and the sample was subsequently heated in the oven for 4 h at 165 °C. After cooling, the Teflon jar was removed from the oven, and the sample was diluted to 10 mL with ultra-pure water for analysis.

The concentrations of Cd, Cr and Pb were determined using an ICP-AES (SPECTRO ARCOS EOP, SPECTRO Analytical Instruments GmbH), and the concentration of As was determined using an ICP-MS (NexION300x, PerkinElmer Instruments Co., Ltd).

One sample replicate was analysed with each batch (twenty samples), and the relative deviation was within 5%. Reagent blanks, and reference materials (GBW 0741, GBW 10047) were also included in each batch of analyses to ensure the quality of the analysis. The recoveries of reference materials for each element ranged between 90% and 110%. The detection limits are 1.2×10^{-5} mg L^{-1}, 0.2×10^{-3} mg L^{-1}, 1×10^{-3} mg L^{-1} and 3×10^{-3} mg L^{-1} for As, Cd, Cr and Pb, respectively. The blank check values were below the detection limits.

2.3. Bioconcentration factor of metal(loid)s

The bioconcentration factor (BCF) indicates the efficiency of the plant in accumulating a metal(loid) into its tissues from soil. The soil-bulb BCFs of metal(loid)s were calculated as follows:

$$BCF = \frac{C_{bulb}}{C_{soil}} \tag{1}$$

where BCF is the bioconcentration factor of the metal(loid) from soil to the edible part of lily, C_{bulb} is the metal(loid) concentration in the bulb of lily in mg kg^{-1}, and C_{soil} is the metal(loid) concentration of the soil in mg kg^{-1} (Ali et al., 2013).