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Accumulation characteristics and potential risk of PAHs in vegetable system grow in home garden under straw burning condition in Jilin, Northeast China

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ABSTRACT

The accumulation characteristics and potential risk posed by polycyclic aromatic hydrocarbons (PAHs) in soils and vegetables grown in the home garden and agricultural field were investigated in this research. The average concentrations of 16 PAHs in soils and vegetables in the home garden were 508.9 ng/g and 197.3 ng/g, respectively, and in agricultural fields were 589.9 ng/g and 171.3 ng/g, respectively. The 16 PAHs concentrations of vegetables in the home garden were a little higher than in agricultural field. The most abundant PAHs in soils and vegetables was Phe, followed by Fla and Pyr in our study area. The concentrations of low-molecular-weight PAHs (L-PAHs) were higher in vegetables as compared to higher molecular weight 4–6 ring PAHs (H-PAHs). The results of plant concentration factor (PCF) indicated that L-PAHs have greater mobility in our research. Based on the results of PAH ratios, the main sources of the PAHs in soils were determined to be the combustion of biomass, coal, and petroleum. The total values of incremental lifetime cancer risk (ILCR) for males and females induced by soils and vegetables in home garden and agricultural field were all about 10^{-7} and 10^{-10}. All the ILCRs value were lower than the baseline value, indicated that the carcinogenic risk for the soils and vegetables contaminated with PAHs in our study area for the residents was negligible.

1. Introduction

Dietary uptake pathway is considered to be the major way of exposure for most people (Wang et al., 2012; Hsu et al., 2009; Oliva et al., 2017), accounting for around 90% intake compared with inhalation or dermal contact (Khillare et al., 2012; Martí-Cid et al., 2008a, 2008b; Martorell et al., 2011; Ferré-Huguet et al., 2008). Some scholars evaluate that about 30% of cancers for people are caused by low exposure to initiating carcinogenic contaminants in the diet (Mansour et al., 2009).

Polycyclic aromatic hydrocarbons (PAHs) as ubiquitous persistent organic pollutions have attracted a lot of attention because of their carcinogenic and mutagenic properties (Yin et al., 2008; Liu et al., 2016a, 2016b; Jiang et al., 2016; Guo et al., 2017; Ahammed et al., 2013; Chen et al., 2018). Crops contaminated by PAHs is a growing problem in the world on account of sewage sludge application, industrial activities, solid waste disposal, wastewater irrigation, and automobiles exhaust (Shi et al., 2005; Khan et al., 2013; Amin et al., 2013; Arora et al., 2008; Silva et al., 2018). The uptakes of contaminants by crop affect the food quality seriously, which bring potential health risk to people (Khillare et al., 2012; Chai et al., 2017).

Soil is the most important environmental reservoir and sinks for PAHs on account of they may enter into the soil through dry and wet deposition from the atmosphere. They are very hard to degrade because of their long half-lives and highly stable chemical structures (Inam et al., 2016; Agarwal, 2009; Li et al., 2011; Nam et al., 2003; Yuan et al., 2015). Investigating the PAHs concentration in soil is the basis to

Abbreviations: PAHs, Polycyclic aromatic hydrocarbons; L-PAHs, Lower molecular weight 2–3 ring PAHs; H-PAHs, Higher molecular weight 4–6 ring PAHs; PCF, Plant concentration factor; ILCR, Incremental Lifetime Cancer Risk; USEPA, United States Environmental Protection Agency; EPA, Environmental Protection Agency; HPLC, High Performance Liquid Chromatography; 7 CarPAHs, 7 Carcinogenic PAHs; GC, Gas Chromatography; TEF, Toxic equivalency factors; SF, Slope factor; ED, Exposure duration; CF, Conversion factor; BW, Body weight; EF, Exposure frequency; AT, Average life span; PEF, Particulate emission factor; $IR_{inhalation}$, Soil inhalation rate; $IR_{ingestion}$, Soil ingestion rate; SA, Dermal surface exposure; AF, Dermal adherence factor; ABS, Dermal adsorption fraction; IRi, Food ingested rate; CSF, Carcinogenic slope factor; CV, Coefficients of variation

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evaluate the pollution levels, emission sources and environmental risk (Wild and Jones, 1995). The continuous accumulation of PAHs in agricultural field soils caused by human activities give rise to potential negative risks both for the agricultural ecosystem and the human through food chain bioaccumulation (Feng et al., 2017; Duan et al., 2015). It is crucial and necessary to study the PAHs concentrations and profiles in soils and vegetables simultaneously and assess the risks. Characteristically, vegetables can take up a lot of essential nutrients along with certain trace elements in a short period. Thus, the safety of vegetables is attracting more attention in recent years (Liu et al., 2005).

Northeast China is one of the important national grain production bases. Due to the relatively backward agricultural production level, many areas, especially in the vast rural areas, also retains the traditional farming methods of burning straw. In the vast rural areas of northeast China, the residents will burn a large amount of straw for plant ash or use it as a living fuel, resulting in environmental pollution such as soil contaminated. In addition, rural residents often use home gardens to grow a variety of vegetables to meet the daily demand for vegetables. And these garden soils and vegetables are exposed to the environment of straw burning and face PAHs pollution potential risks. A large number of studies have been focused on the contamination of PAHs in farmland soils, garden soils of urban, vegetables grown in urban areas and vegetables grown in farmland. But, there is nearly no research conducted regarding the potential risk of PAHs in vegetable system grown in home garden. Therefore, to ensure the human health and diet safety of residents, it is essential to investigate the accumulation condition and potential risk of PAHs in soil-vegetable system under straw burning condition. The objective of this study is to determine the level of PAHs contamination in soils and vegetables under straw burning condition and evaluate the health risk for the residents.

2. Materials and methods

2.1. Sample collection

The study area is located at the northwest of Jilin province (123°25′–124°23′E, 44°37′–45°18′N), the middle area in Songnen plain (Fig. 1), covers about 3610 km^2 and has a total population of 30 million. The area is in a warm temperate zone with a monsoon climate and belongs to the arid and semi-arid region. The mean annual air temperature is about 4.6 °C and the mean annual precipitation is 420.6 mm. The main soil type is light chernozem soil.

2.2. Sample collection

51 vegetable samples including 37 leafy vegetables (6 lettuces, 7 cabbages, and 10 Chinese cabbages in the home garden; 5 cabbages and 9 Chinese cabbages in farmland), 14 tuber vegetables (5 carrots in the home garden and 9 carrots in farmland) were collected in September, 2016. Only edible parts were sampled for analyses. 51 soil samples were collected from the same sites where vegetable samples were collected with a pre-cleaned stainless steel soil auger.

Vegetables were washed with tap water to remove all visible soil particles, freeze-dried, ground to pass through a 2 mm sieve and stored at − 20 °C. All the parts of each vegetable were analyzed to measure the concentrations of PAHs in triplicates. Soil samples were air-dried and ground to pass through 2 mm mesh and stored at − 20 °C in paper sample bag until further analyses. All the soil samples were analyzed in triplicates.

2.3. Chemicals

Standard mixture containing 16 United States Environmental Protection Agency (USEPA) priority PAHs (16 compounds specified in Environmental Protection Agency (EPA) method 610, each at 100 mg/L) were obtained from the National Standard Material Center (Beijing, China). All solvents (dichloromethane, acetone, n-hexane, etc.) were supplied by Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China), used for sample extraction and purification were of High Performance Liquid Chromatography (HPLC) grade. Copper power and anhydrous sodium sulfate were obtained from Sinopharm Chemical Reagent Co., Ltd.

2.4. Extraction

For each sample, 10 g vegetable or 5 g soil was mixed with 5 g of copper power to move off sulfur, and extracted in ultrasonic bath with 40 mL of dichloromethane: acetone (1:1/v:v) for 15 min (repeated three times). The details of the laboratory analyses of vegetable and soil were the same as described in our previous research (Chen et al., 2016). There are 16 PAH compounds recommended as priority by USEPA, including 7 carcinogenic PAHs (7 carPAHs) and 9 non-carcinogenic PAHs (Chen et al., 2018). They were measured by gas chromatography (GC) equipped with a flame ionization detector (Clarus 680, PerkinElmer Inc., Waltham, MA, USA). An HP-5 capillary column (30 m, 0.25 mm inner diameter 0.25 µm film thickness, Agilent, USA) was used for chromatographic separation. Helium (99.999%) was used as a carrier gas, and set at a constant flow of 1 mL/min. The GC oven temperature was initially held at 50 °C for 2 min and programmed to increase to 200 °C (19 °C/min) for 2 min. Then, the oven temperature was programmed to increase to 240 °C (4.5 °C/min) and held for 2 min, and finally increased to 300 °C (2.5 °C/min) and held for 5 min.

2.5. Quality control

The method blank was applied, and the sample duplicate was analyzed for quality control and assurance. The standard external method was used with a 16 PAHs reference material mixture, and the calibration curves were linear in the concentration range with linear regression coefficients (R^2) > 0.99. Sample contamination was avoided by use of cleaned glassware, analytical grade chemicals and high-purity deionized water. The recoveries of matrix spikes for vegetable samples ranged from 67% to 101%. For soil samples, the recoveries ranged from 74% to 125%. Method blanks operation of laboratory showed no detectable amounts of PAHs. All results are expressed on a dry weight basis.

2.6. Transfer factor

The plant concentration factor (PCF) of PAHs from soil to the vegetable was computed as the ratio of PAHs concentration in edible parts of vegetables with the PAHs concentration in soils. The PCF was calculated by the following equation (Rehman et al., 2017):

$$PCF = C_{vegetable}/C_{soil} \tag{1}$$

where, $C_{vegetable}$ and C_{soil} represent the PAHs concentrations in vegetable and soil on dry weight basis, respectively.

2.7. Risk assessment

The incremental lifetime cancer risk (ILCR) was used to quantitatively estimate the exposure risk for environmental PAHs based on standard models (USEPA, 1991). The population of our study area was divided into six groups according to age and gender: childhood (0–10 years) male and female, adolescence (11–18 years) male and female, and adulthood (19–70 years) male and female (Hu et al., 2017)

The following formulas were used to evaluate the ILCR in term of ingestion, dermal contact and inhalation. The parameters used to determine the cancer risk of soil in these models are defined in Table 1(Wang et al., 2017).