



Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Characterization of polycyclic aromatic hydrocarbons (PAHs) in vegetables near industrial areas of Shanghai, China: Sources, exposure, and cancer risk

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ARTICLE INFO

Article history:

Received 2 March 2018

Received in revised form

22 May 2018

Accepted 1 June 2018

Keywords:

PAHs

Dietary exposure

Cancer risk

Vegetable contamination

Industrial areas

ABSTRACT

Dietary consumption of contaminated vegetables may contribute to polycyclic aromatic hydrocarbon (PAH) exposure in humans; however, this exposure pathway has not been examined thoroughly. This study aims to characterize the concentrations of PAHs in six types of vegetables grown near industrial facilities in Shanghai, China. We analyzed 16 individual PAHs on the US EPA priority list, and the total concentration in vegetables ranged from 65.7 to 458.0 ng g⁻¹ in the following order: leafy vegetables (romaine lettuce, Chinese cabbage and Shanghai green cabbage) > stem vegetables (lettuce) > seed and pod vegetables (broad bean) > rhizome vegetables (daikon). Vegetable species, wind direction, and local anthropogenic emissions were determinants of PAH concentrations in the edible part of the vegetable. Using isomer ratios and principal component analysis, PAHs in the vegetables were determined to be mainly from coal and wood combustion. The sources of PAHs in the six types of vegetables varied. Daily ingestion of PAHs due to dietary consumption of these vegetables ranged from 0.71 to 14.06 ng d⁻¹ kg⁻¹, with contributions from Chinese cabbage > broad bean > romaine > Shanghai green cabbage > lettuce > daikon. The daily intake doses adjusted by body weight in children were higher than those in teenagers and adults. Moreover, in adults, higher concentrations of PAHs were found in females than in males. For individuals of different age and gender, the incremental lifetime cancer risks (ILCRs) from consuming these six vegetables ranged from 4.47 × 10⁻⁷ to 6.39 × 10⁻⁵. Most were higher than the acceptable risk level of 1 × 10⁻⁶. Our findings demonstrate that planting vegetables near industrial facilities may pose potential cancer risks to those who consume the vegetables.

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1. Introduction

PAHs are generated during incomplete combustion of carbon-containing substances, such as wood, coal, gasoline, and diesel (Peng et al., 2011; Shen et al., 2011). The carcinogenicity of PAHs is now well established (Boström et al., 2002; Menzie et al., 1992). PAHs are widely distributed in various environmental media, including the atmosphere, water, sediment, and soil. They are taken up by plants due to their lipophilic and hydrophobic properties.

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Dietary consumption is a major route of exposure for PAHs, which can account for >70% of total exposure in non-smoking individuals (Khillare et al., 2012; Martorell et al., 2011; Wang et al., 2018). Vegetables are an important part of the human diet and important sources of nutrients and vitamins (Camargo and Toledo, 2003; Wang et al., 2018). Vegetables can be contaminated with PAHs through atmospheric deposition, irrigation, and soil uptake.

Numerous studies have characterized PAHs in the atmosphere and soil and have associated PAHs with health risks (Niu et al., 2017; Shi and Zhao, 2014; Zhang et al., 2008). However, few studies have considered the exposure pathway of vegetable consumption, despite the fact that PAHs in atmosphere and soil can accumulate in vegetables. Therefore, it is important to assess the health risk of PAHs in vegetables exposed to air and soil that may be

polluted by PAHs, such as vegetables grown near industrial areas. Fismes et al. (2002) studied the migration and transformation of PAHs from the soil to vegetable roots and found that the concentrations of PAHs in vegetables increased with increasing soil PAH concentrations. Atmospheric absorption is another reported source of PAHs in vegetables. Kipopoulou et al. (1999) found that the concentrations of PAHs in the above ground parts of plants were greater than those of the underground parts. Wang et al. (2011) also found that lower molecular weight PAHs were found in higher proportions in the roots and shoots of vegetables than higher molecular weight PAHs, and atmospheric absorption was the main accumulation route.

In this study, we examined PAHs in vegetables grown near industrial facilities in suburban areas of Shanghai. Shanghai is one of the most highly developed and densely populated cities in China. Specifically, our study had the following objectives: (1) measure PAH concentrations in a variety of vegetables growing on farmland near industrial areas; (2) explore the factors that could affect the accumulation of PAHs in vegetables; (3) identify the possible sources of PAHs in different vegetables; and (4) evaluate the potential risk to human health through the dietary intake of vegetables growing in polluted areas. The results indicate PAH contamination in vegetables is a serious concern for consumers, and efforts should be made to improve urban land resources, food security and human health related to vegetable cultivation near industrial facilities.

2. Materials and methods

2.1. Study area

There were 3.15×10^5 ha (ha) of agricultural land in Shanghai suburbs, which covered 37.7% of the total land area of Shanghai (Shanghai Municipal Planning and Land & Resources Administration). Local residents often grow vegetables on agricultural land near industrial areas, making the consumption of locally grown vegetables a potential exposure route for PAHs. We selected seven agricultural sites near the Wujing chemical industry area, Shanghai Baoshan Iron and Steel Plant, Yuqiao Municipal Waste Incineration Power Plant and highways. Detailed information regarding the sampling is described by Bi et al. (2018). In brief, sites MH-1, MH-2, MH3 and MH4 are located southeast, west and south of Wujing chemical industry area, respectively. Site BS is located to the west of Shanghai Baoshan Iron and Steel Plant and adjoins a trunk route. Site PD is located to the south of Yuqiao Municipal Waste Incineration Power Plant. Site FX adjoins the Hujin highway. Site QP is relatively far from any large industrial facilities and is mainly used to grow strawberries and vegetables in a suburban area of west Shanghai. Therefore, we used this site as a control (Fig. 1).

2.2. Sampling and preparation

According to the growth characteristics of different vegetables, six categories of commonly grown and consumed vegetables were selected. Leafy vegetables, including Chinese cabbage (*Brassica rapa* var.), Shanghai green cabbage (*Brassica chinensis* var.), romaine (*Spinacia oleracea* var.) and rhizome vegetables, such as daikon (*Raphanus sativus* L.), were collected in January 2014. The broad bean (*Vicia faba* L.), which belongs to seed and pod vegetables and lettuce (*Lactuca sativa* L.), belonging to stem vegetables, were collected in May 2014. Composite samples were generated from 3 to 5 plants of each species. The fresh vegetable samples were washed with tap water and rinsed with deionized water. The fresh weight was recorded after air-drying. After being dried at -54°C in a lyophilizer (Christ, Germany), the samples were ground using a

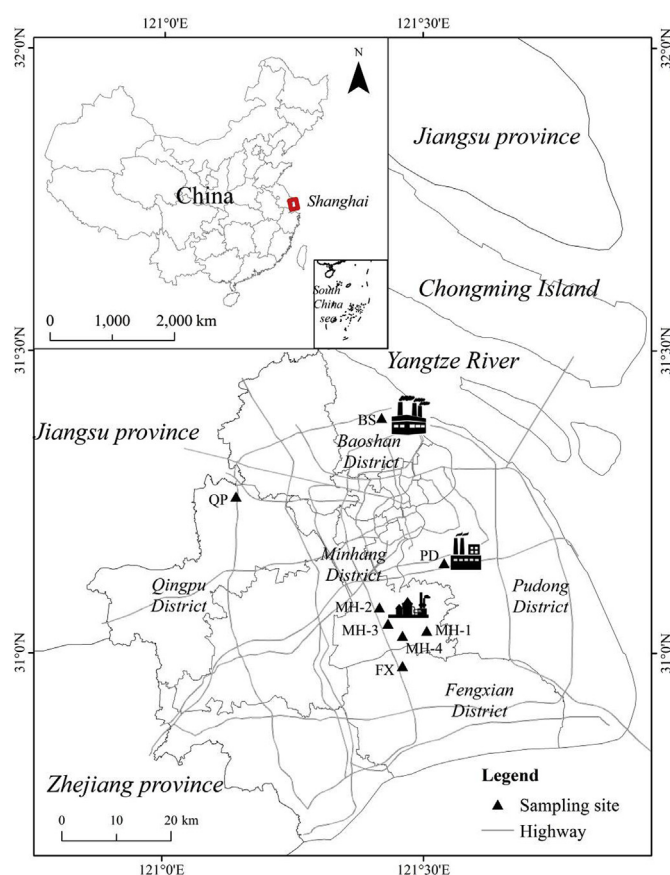


Fig. 1. Sampling sites of vegetables in Shanghai.

mortar and filtered through a $250\ \mu\text{m}$ mesh sieve. The resulting particles were refrigerated at -18°C until analysis.

Pesticide grade reagents (dichloromethane, acetone and *n*-hexane) were obtained from Merck (Germany). Neutral aluminum oxide, granular anhydrous sodium sulfate and quartz sands were obtained from Sinopharm Chemical Reagent Co., Ltd (SCRC). Decachlorobiphenyl and deuterated PAHs (naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12) were purchased from Dr. Ehrenstorfer (Germany). After ultrasonic cleaning, all glassware was dried at 450°C for 4 h. Silica gel (100–140 mesh) was activated at 130°C for 16 h and other reagents, including glass wool (CNW, USA), alumina and quartz sands were heated at 450°C for 4 h.

2.3. Sample analysis

Samples were extracted using accelerated solvent extraction (ASE300, Dionex, USA). Extractions were performed under 1500 psi and 120°C for a 6 min static cycle. The extraction was purified using a silica-aluminum column. The eluate was concentrated by DryVap automatic concentrator (Horizon, USA). The PAH concentrations in the samples were analyzed by a gas chromatograph coupled with mass spectrometry (GC/MS) (7890A/5975C, Agilent Technology, USA) and equipped with a HP-5MS capillary column ($30.0\ \text{m} \times 0.25\ \text{mm} \times 0.25\ \mu\text{m}$). The concentrations of sixteen PAHs were determined: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[k]fluoranthene (BkF), benzo[b]fluoranthene (BbF), benzo[a]pyrene (Bap), indeno[123cd]pyrene (Icdp), dibenz

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