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Original Research

Accumulation and Health Risk Assessment of PAHs in Radish

Jin Guo[1], Chao Chai[1]*, Wei Ge[2], Lusheng Zeng[1], Juan Wu[1], Dan Xiang[1], Xiaomei Zhang[1]

[1]Qingdao Engineering Research Center for Rural Environment, Qingdao Agricultural University, Qingdao, China
[2]College of Life Sciences, Qingdao Agricultural University, Qingdao, China

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Abstract

Three kinds of soil with different levels of polycyclic aromatic hydrocarbon (PAH) contamination were collected, and diesel was added to soil to prepare two kinds of diesel-contaminated soil. The radish was planted in five kinds of soil through a pot experiment, and the concentration and composition of PAHs in radish were analyzed using high-performance liquid chromatography with an ultraviolet and fluorescent detector. The PAH contribution in aboveground parts of radish from atmospherically deposited particulates was studied, and the health risk of ingesting contaminated radish was assessed. Results showed that PAH concentrations (196.2-982.6 ng/g) in the parts of radish found underground were significantly higher than in aboveground parts (129.7-556.7 ng/g, $p<0.05$). Predominant PAH compounds in radish were the 3- and 4-ring PAHs, accounting for 78.1-92.7%. In general, the values of root concentration factors (RCFs: 0.30-0.55) were significantly higher than shoot concentration factors (SCFs: 0.19-0.39, $p<0.05$). Atmospherically deposited particles contributed less than 1% of the PAHs in aboveground parts of radish, which indicated two things: the atmospheric particles had a slight effect on the PAH content in aboveground parts of radish , and the soil contributed more to PAH accumulation in aboveground parts of radish than the particles. The total toxicity equivalence quotient in radish grown in diesel-contaminated soil samples was higher than in other types of soil. Ingestion of radish planted in five kinds of soil had no carcinogenic risk to children, adolescents, and seniors; whereas ingestion of radish from heavily contaminated and diesel-contaminated soil samples had carcinogenic risks to adults.

This study highlights the accumulation and potential health risks associated with cultivation and consumption of radish in soil with different contamination levels and sources of PAHs.

Keywords: PAHs, accumulation, health risk assessment, radish

*e-mail: chaichao1999@126.com

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are composed of two or more aromatic rings of carbon and hydrogen atoms and are considered a large class of ubiquitous persistent organic pollutants in the environment [1-3]. PAHs mainly come from burning coal and fossil fuel, industrial and vehicle emissions, petroleum spill petroleum chemical industry, coking, and smelting [4]. PAHs drew concern for their genotoxic and carcinogenic potential [5-7]. The U.S. Environmental Protection Agency (USEPA) has classified 16 priority PAHs, seven of which are potential carcinogens [8].

Many studies have shown that vegetables can accumulate PAHs from polluted environments, such as in contaminated soil, as reported by Zhang et al. [9]. The concentration of PAHs in lettuce, potato, and carrot increased with the rising concentrations in soils [10]. PAHs mainly accumulate in a pathway from the soil to the root and shoot of vegetables. Yang et al. [11] also suggested that phenanthrene accumulation in the shoot mainly comes from root transport. The main factors affecting PAH uptake by plants were the initial PAH concentration in soil, plant species, and soil microbial community. However, Wan et al. [12] showed that the concentration of PAHs in 77 kinds of vegetables and in soils are not significantly correlated. PAH concentration in plant aerial tissue is positively correlated with the concentration in air; PAHs in atmospheric vapor may be an important source of PAHs in five vegetable species [13]. PAHs in vegetables may be harmful to human health because the main route of human exposure to environmental pollutants is food consumption, which accounts for 90% of intake compared to inhalation and skin contact [14-16]. Therefore, studying PAH accumulation in vegetables is important.

Most studies previously conducted on plant PAH uptake and accumulation focused on leafy vegetables [17-18]. Waqas et al. [19] reported that cabbages, followed by radishes in Mardan, had the highest concentration of PAHs. These results indicate that PAH accumulation amounts in root vegetables is higher than in leafy vegetables due to wastewater irrigation. A few studies have studied the effect of atmospherically deposited particulates on PAH accumulation in root vegetables; thus, more attention should be provided for this aspect. Although studies carried out so far on health risk assessment of PAHs in vegetables grown in agricultural soils contaminated by coal and wood combustion are many [20-21], few studies have investigated the health risk assessment of PAHs in edible vegetables grown in diesel-contaminated soils. Diesel is widely used as one of the most vital petroleum products and contains paraffins, olefins, naphthenes, PAHs, and other groups [22]. About 800 t petroleum are released annually into environments in the world, leading to the contamination of PAHs in soils [23]. Therefore, the health risk assessment of PAHs in vegetables grown in diesel-contaminated soils should be of concern.

Radish (*Raphanus sativus* L.) belongs to the family of *Brassicaceae* and presents high nutritional and pharmaceutical value. Many countries have cultivated varieties of radish, one of the most common vegetables; hence, radish was selected as the representative root vegetable for this study. This study aimed to investigate the concentration and profile of PAHs in radish planted in soil with different contamination levels and sources of PAHs and analyze the relationship among PAH concentrations of radish, soils, and atmospherically deposited particulates. A discussion of the contribution of PAHs to radish of atmospherically deposited particulates and an assessment of the health risk of PAHs follow. The results of the study may be very helpful in elucidating accumulation of PAHs in root vegetables and protecting residents from the potential health risks through vegetable ingestion caused by different PAH contamination sources – especially diesel contamination.

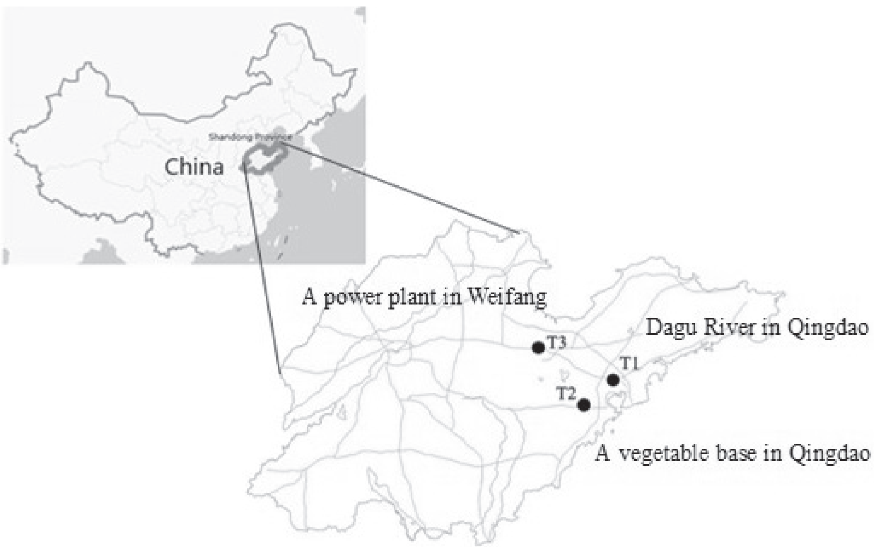

Fig. 1. Schematic map showing the soil sampling sites.

Materials and Methods

Collecting and Preparing Soils

The soils were collected in 2016 in Shandong Province from three sites, including along the Dagu River in Qingdao (T1, Fluvo-aquic soils), at a vegetable base in Qingdao (T2, Brown earths), and from an agricultural field near a power plant in Weifang (T3, Brown earths; Fig. 1.). The pH values of T1, T2, and T3 soils were 6.1, 5.8, and 6.0, respectively, and the concentrations of organic matter were 5.3, 15.7, and 16.9 g/kg, respectively. Varying amounts of diesel were added to soil T1 to prepare diesel-contaminated soils T4 and T5. The proportions of diesel in T4 and T5 soils were approximately 10 mL/kg and 15 mL/kg, respectively. Then, T4 and T5 soils were aged for eight weeks. Soils were air dried in the dark and then sieved through a 2 mm mesh for a pot experiment.

Design of Pot Experiment

Radish seeds were planted in T1-T5 soils. Each pot was filled with 3.5 kg of soil (<2 mm), and four replicates per treatment were prepared. Deionized water was used for irrigation throughout the entire process, which lasted 35 days. The atmospherically deposited particles were collected with silica gel placed at shoot height to analyze the PAH contribution of atmospherically deposited particles to shoot tissues [24]. The soils were covered with uncontaminated sand to prevent possible root contact with atmospheric deposition. After 35 days, the soil and plant samples were collected, freeze-dried, and ground to determine PAH concentrations.

PAH Extraction and Analysis

Two grams of soil and 10 mL dichloromethane (DCM) were placed in the centrifuge tube and sonicated for one hour. Then the mixture was centrifuged at 4,000 rpm for 10 minutes. The same extraction process was repeated. The extracts were combined and concentrated to 1 mL. The solvent underwent purification through silica gel column (the upper was 4 g anhydrous sodium sulfate, the lower was 4 g silica gel) with elution of hexane and DCM (11 mL, v:v, 1:1). The eluent was concentrated to near dryness and solvent-exchanged to 2.0 mL methanol for analysis. A wash with 10 mL of DCM in the glass centrifuge tube removed atmospherically deposited particles. This method was consistently used with soil.

Plant samples (2 g) were extracted through ultrasonication for 10 min with a solution of acetone and hexane (v:v, 1:1). The process was repeated thrice and centrifuged at 2,500 r/min for 5 min. The solvent was purified through acidic silica gel column (the upper was 4 g anhydrous sodium sulfate, the middle was 4 g sulfuric acid silica gel, the lower was 4 g silica gel) with 20 mL of 1:1 (v/v) elution of hexane and DCM. The eluent was concentrated to near dryness and solvent-exchanged to 2.0 mL methanol for analysis.

PAHs were analyzed using high-performance liquid chromatography equipped with an ultraviolet and fluorescent detector (HPLC–UV–FLD, Shimadzu). An Inertsil ODS-P column (250 × 4.6 mm, 3.5 μm particle size, 1,000 nm pore size) was used to separate 16 PAHs. The mobile phase was methanol-water (80:20, v:v), and the flow rate was 1.0 mL/min. Column temperature was 40°C and the injection volume was 20 μL.

A composite standard solution of 16 PAHs, including naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenzo(a,h)anthracene (DBA), indeno(1,2,3-cd)pyrene (IPY), and benzo(ghi)perylene (BPE) was purchased from AccuStandard Company (USA). All solvents were HPLC grade.

Quality Assurance and Control

Data quality was controlled by blank experiment and parallel sample analysis. No detectable amount of PAHs was found in the blank operation. The recovery of NAP was 62%, and the other PAHs were 86%±15%. Quantitative analysis was performed using a five-point calibration curve method. The correlation coefficients of each calibration curve were higher than 0.999. The standard mixture was analyzed for every 10 samples to determine instrument stability and confirm the calibration curve. The limits of detection were in the range of 0.07 ng/g to 2 ng/g.

Data Analysis

Shoot and Root Transfer Factors

The soil-to-plant transfer is one of the main ways in which pollutants enter the food chain. Root concentration factors (RCFs) and shoot concentration factors (SCFs) were calculated as follows:

$$\text{(i)} \quad RCF = \frac{C_{underground}}{C_{soil}} \quad (1)$$

$$\text{(ii)} \quad SCF = \frac{C_{aboveground}}{C_{soil}} \quad (2)$$

…where $C_{underground}$, $C_{aboveground}$, and C_{soil} represent PAH concentrations in the underground parts of radish, aboveground parts of radish, and soil based on dry weight, respectively [24].

Contributions of PAHs in Atmospherically Deposited Particulates

The radish leaf surface area and mass of atmospherically deposited particulates were calculated by Fismes et al. [11]. The contribution of PAHs in aboveground parts of radish from atmospherically deposited particulates was calculated as follows:

$$Dd = SA \times AdDd \quad (3)$$

…where Dd is the daily dust deposition quality on aboveground parts of radish (mg), SA is the surface area of the radish leaf (m^2), and AdDd is average daily dust deposition (mg/m^2).

$$TAd = Dd3 + \frac{Dd5}{2} \times 35d \quad (4)$$

…where TAd, Dd3, and Dd5 represent the total average dry dust deposition on aboveground parts of radish (mg) and daily dust deposition on aboveground parts of radish (mg) at 3 and 5 weeks, respectively.

$$ATd - tPAHs = C - tPAHs \times TAd \times 10^{-6} \quad (5)$$

…where ATd − tPAHs is the average total deposition of the total PAHs (μg) on aboveground radish parts, and C − tPAHs represents the average total PAH concentrations in the dust (ng/g) [24].

$$Contribution = \frac{ATd\text{ - }tPAHs}{C_{aboveground} \times M_{aboveground}} \quad (6)$$

…where $M_{aboveground}$ represents the mass of aboveground parts of radish based on dry weight.

Health Risk Assessment

Toxicity equivalence quotient (TEQ_{BaP}) was shown as BaP equivalent concentrations and was calculated by multiplying the concentration of each PAH in the radish by its TEF based on the USEPA [25].

$$TEQ_{Bap} = C_i \times TEF_i \quad (7)$$

…where C_i is the concentration of individual PAH, and TEF_i is the corresponding toxicity equivalence factor.

The incremental lifetime cancer risk (ILCR) of the dietary exposure to PAHs was also calculated based on Equation 8 [26].

$$ILCR = TEF_{Bap} \times E_f \times IR \times EF \times ED \times SF \times CF/(BW \times AT) \quad (8)$$

…where E_f, a conversion factor of 0.009, was the weight of fresh radish converted to dry weight; IRs (amount of radish ingested) of children, adolescents, adults, and seniors were 0.0043, 0.0072, 0.011, and 0.010 kg/d, respectively [27]; SF was the oral cancer slope factor, BaP (7.3 mg/kg/d) [28]; ED factors (duration of exposure) of children, adolescents, adults, and seniors were 7, 7, 43, and 10, respectively [26, 29]; BWs (average body weight) of children, adolescents, adults, and seniors, were 23.6, 48, 62, and 62 kg, respectively; AT or average time was 27,740 days [30]; EF or exposure frequency was 365 days/year; and CF is the conversion factor (10^{-6} mg/ng).

Data Analysis

The analysis was conducted through SPSS version 20.0 using ANOVA with the least significant difference method (at $\alpha \leq 0.05$ level) to calculate the significant statistical difference in PAH concentrations. The correlation analysis was carried out using the Spearman correlation analysis.

Result and Discussion

Concentration and Profile of PAHs in Soils

The concentrations and relative contributions of PAHs in soils are shown in Table 1. The total concentra-tions of the 16 PAHs (Σ_{16}PAHs) ranged from 376.7-2,509.1 ng/g in soils. Σ_{16}PAHs in T3 soil was the highest, and Σ_{16}PAHs in the four other soils were below 1,000 ng/g. Relative to the standard of PAH concentration in soils [31], T1 soil was weakly contaminated (200-600 ng/g); T2, T4, and T5 soils were contaminated (600-1,000 ng/g); and T3 soil was heavily contaminated (>1,000 ng/g).

The profile of PAHs in the five soil samples was slightly different. In T1 soil, PYR, ACE, and ACY were the principal compounds. Chief compounds in T2 soil were ACY, FLA, and PYR. Meanwhile, PHE and FLU were predominant in T3 soil, and ACE, ACY, and ANT were predominant in T4 and T5 soils. Despite the difference in profiles, the contributions of 3- and 4-ring PAHs were relatively high in all soils. The contributions of 3-ring PAHs in T1, T3, T4, and T5 soil accounted for 42.7%, 75.2%, 47.6%, and 51.8% of the Σ_{16}PAHs, respectively, whereas the contribution of 4-ring PAHs in T2 soils accounted for 46.3% of the Σ_{16}PAHs.

Σ_{16}PAHs in T2 and T4 soil were roughly similar, but the profiles of PAH in T2 and T4 soil were slightly different. The contribution of BkF, BaP, BPE, and IPY in T4 soil was higher than T2 soil, thereby suggesting that the contributions of 5- and 6-ring PAHs in T4 soil were higher than T2 soil. The PAH profiles in diesel-contaminated soils of T4 and T5 were similar. The contribution of BaP in T4 (8.8%) and T5 (8.7%) was higher than T1 (4.6%), T2 (2.0%), and T3 (1.4%) soil, thereby suggesting that the contribution of 5-ring PAHs in T4 and T5 soils was higher than T1, T2, and T3. Pan et al. [32] also reported that the concentration of CHR or BaP was high in diesel-contaminated soils.

The concentration of carcinogenic PAHs (Σ_7PAH, including BaA, CHR, BbF, BaP, BkF, DBA, and IPY) [33] ranged 107.1-315.4 ng/g, and Σ_7PAHs in diesel-contaminated soils of T4 and T5 were higher than those in other soils. In this study, diesel was added to T1 soil to prepare the diesel-contaminated soils of T4 and T5, and the contribution of carcinogenic PAHs in T4 (36.9%) and T5 (32.9%) soil were higher than T1 (28.4%) soil. Similar results were reported in previous studies. Qi et al. [34] reported that the concentrations of carcinogenic PAHs ranged 47,033.2-287,922.3 ng/g in petroleum-contaminated soils of Shengli Oil Field, and the contributions of carcinogenic PAHs in petroleum-contaminated soils (75.9-78.6%) were high.

PAH Concentrations in Radish

Σ_{16}PAHs in radish are shown in Fig. 2. Σ_{16}PAHs found in buried parts of radish planted in five soils ranged 196.2-982.6 ng/g, while those in parts aboveground ranged from 129.7-556.7 ng/g. Σ_{16}PAHs in radish planted in T3 soil were the highest, whereas T1 soil had the lowest concentration. Plant uptake of PAHs was via fine roots [35], and there are a lot of fine roots in buried parts of radish. Σ_{16}PAHs in parts found underground were significantly higher than those in parts aboveground ($p<0.05$). These results are consistent with those obtained from previous studies [36-37].

Σ_{16}PAHs in radish had a significantly positive correlation with those in soil ($p<0.01$). The correlation coefficients between the two parts of the radish and the soil were 0.976 and 0.956, respectively. The correlation coefficient between the parts of radish found aboveground and underground was 0.956. These results are consistent with those obtained from previous studies. The uptake and

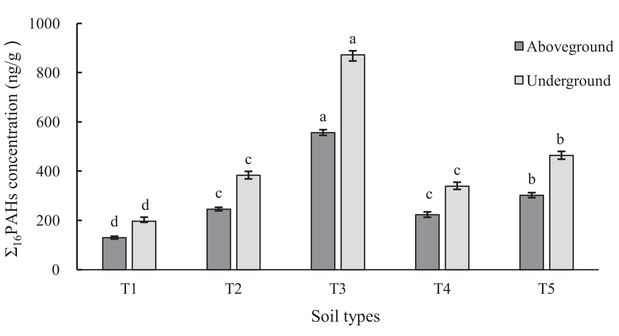

Fig. 2. Σ_{16}PAHs in radish planted in different soils (letters indicate significant differences between the different groups ($p<0.05$), and the same letters show the insignificant difference).

accumulation of PHE, PLA, and BaP by pakchoi cabbage was reported, and the concentrations in roots or shoots increased as PAH concentrations in soil increased [38]. Σ_{16}PAHs in four kinds of leafy vegetables were positively correlated with PAH concentrations in soil [39]. Shen et al. [40] reported that Σ_{16}PAHs in plants are positively correlated with soil and air concentrations and posited that Σ_{16}PAHs in the air of the sampling area were higher (4,050.0 ng/g) than in the soil (724.0 ng/g). However, Zhang et al. [41] found that Σ_{16}PAHs in lettuce, cucumber, cabbage, and other vegetables were not correlated to those in the soil possibly due to the low PAH concentration in soil (233.0 ng/g). The contamination level of PAHs in soils may affect the relationship of PAH concentrations in vegetables and soils.

Compared with Σ_{16}PAHs in soils in Table 1, Σ_{16}PAHs in radish were significantly lower than those in the soil ($p<0.01$). Khana et al. reported that low molecular weight PAH (LMW-PAHs) concentrations in the roots and leaves of lettuce was two to three times and four to five times lower than those in soils, respectively [24]. Meanwhile, high molecular weight PAH (HMW-PAHs) concentrations in roots and leaves were two to three times and 10 to 16 times lower than those in soils.

Profile of PAHs in Radish

The relative contributions of 3- and 4-ring PAHs were high in radish planted in five soils (Fig. 3) with underground portions contributing 81.0-92.7% and parts aboveground contributing 78.1-91.1%. The contribution of 4-ring PAHs was high in radish planted in T2 soil, and the contribution of 3-ring PAHs was high in radish planted in T1, T3, T4, and T5 soils; whereas the contributions of 5- and 6-ring PAHs were relatively low at 3-19%. Previous studies found that the contributions of 2- and 3-ring PAHs in the roots and leaves of winter wheat on oily sludge-amended soil were 57.8% and 53.5%, respectively [42]. In addition, the contributions of 3- and 4-ring PAHs in pine needles ranged from 65-95% [43]. Gao et al. [44] also reported that the contributions of 3-ring PAHs in carrots, spinach, and eggplant were 65.9%, 66.2%, and 64.5%, respectively. Therefore, 2–4-ring PAHs are dominant in vegetables and other plants.

Because of the high contribution of BaP in diesel-contaminated soils of T4 and T5, the contributions of BaP in radish planted in diesel-contaminated soils of T4 and T5 is high. PAH profiles of T2 and T4 soils were slightly different, and the contributions of 5- and 6-ring PAHs in radish planted in T4 soil were higher than T2 soil. Therefore, the contributions of PAHs in radish were similar to those in soil.

A significant positive correlation exists between 2-, 3-, and 4-ring PAHs in radish and those in soils ($r>0.940$, $p < 0.05$), whereas no significant correlation exists between 5-and 6-ring PAHs in radish and those in soils ($p>0.05$). Zhang et al. [45] found that PAHs in soils affected PAHs in vegetables, and a highly similar distribution of PAHs

Table 1. PAH concentrations (ng/g) and relative contributions of individual PAH compounds or of PAHs with different numbers of rings to total PAHs (%).

PAHs	T1		T2		T3		T4		T5	
	Concentration	Contribution	Concentration	Contribution	Concentration	Contribution	Concentration	Contribution	Concentration	Contribution
NAP	1.9	0.5	26.9	3.8	32.9	1.3	10.2	1.4	15.8	1.6
ACY	39.0	10.4	190.9	26.6	54.3	2.2	100.7	14.2	159.8	16.7
ACE	40.8	10.8	30.4	4.2	305.5	12.2	81.8	11.5	115.3	12.0
FLU	27.5	7.3	52.1	7.3	371.5	14.8	34.5	4.9	68.6	7.2
PHE	26.5	7.0	19.8	2.8	847.9	33.8	27.2	3.8	42.5	4.4
ANT	27.1	7.2	4.4	0.6	306.6	12.2	93.4	13.2	110.9	11.6
FLA	33.5	8.9	119.4	16.7	144.9	5.8	27.5	3.9	46.1	4.8
PYR	48.5	12.9	103.9	14.5	255.3	10.2	50.8	7.2	53.1	5.5
BaA	2.5	0.7	60.3	8.4	37.7	1.5	53.7	7.6	59.0	6.2
CHR	22.8	6.1	47.9	6.7	32.7	1.3	47.6	6.7	51.4	5.4
BbF	26.7	7.1	29.7	4.1	37.7	1.5	41.4	5.8	38.3	4.0
BkF	10.0	2.7	4.5	0.6	18.6	0.7	22.6	3.2	40.5	4.2
BaP	17.3	4.6	14.4	2.0	35.6	1.4	62.7	8.8	83.4	8.7
DBA	3.7	1.0	0.9	0.1	0.8	0.0	2.6	0.4	4.3	0.4
BPE	24.8	6.6	6.3	0.9	17.4	0.7	21.7	3.1	31.3	3.3
IPY	24.1	6.4	4.8	0.7	9.7	0.4	31.1	4.4	38.5	4.0
2-ring PAHs	1.9	0.5	26.9	3.8	32.9	1.3	10.2	1.4	15.8	1.6
3-ring PAHs	160.9	42.7	297.4	41.5	1,885.7	75.2	337.6	47.6	497.1	51.8
4-ring PAHs	107.3	28.5	331.5	46.3	470.7	18.8	179.6	25.3	209.7	21.9
5-ring PAHs	57.6	15.3	49.5	6.9	92.8	3.7	129.3	18.2	166.6	17.4
6-ring PAHs	48.9	13.0	11.2	1.6	27.2	1.1	52.8	7.4	69.8	7.3
Σ_7PAH	107.1	28.4	162.5	22.6	172.8	6.8	261.7	36.9	315.4	32.9
Σ_{16}PAH	376.7	100.0	716.6	100.0	2,509.1	100.0	709.5	100.0	958.8	100.0

in vegetables and soils was observed. The significant contributions of 3- and 4-ring PAHs in five soils in this study led to a large proportion of 3- and 4-ring PAHs in radish. LMW-PAHs more easily accumulate due to their considerable vapor pressure, water solubility, and bioavailability [46-48]. PAH accumulation was reduced with the increase of the number of benzene rings in molecular PAHs [49]. Physicochemical properties of the soil are factors affecting PAH profiles in vegetables. Zhang et al. [50] found that the high concentration of 3-ring PAHs in vegetables was related to their responsive interaction with fulvic acid, the high concentration of 3-ring PAHs in soil, and high bio-accessibility.

The average proportion of carcinogenic PAHs in radish ranged 15.6-30.7%. The average proportion of carcinogenic PAHs in radish planted in T4 soil (30.7%) and T5 soil (26.6%) was higher than in radish planted in T1, T2, and T3 soils due to the higher proportion of carcinogenic PAHs in T4 and T5 soils (Table 1). Khillare et al. [51] reported that carcinogenic PAHs accounted for 8% and 11% of total PAHs in spinach and radish planted in the soils around thermal power plants, respectively. Wang et al. [52] found that the proportions of carcinogenic PAHs were 13.9% and 7.5% in Chinese cabbage and Indian lettuce planted in the agricultural soils, respectively. Therefore, the proportion of carcinogenic PAHs in vegetables grown in diesel-contaminated soil is higher than that in other soils due to the higher proportion of carcinogenic PAHs in diesel-contaminated soil.

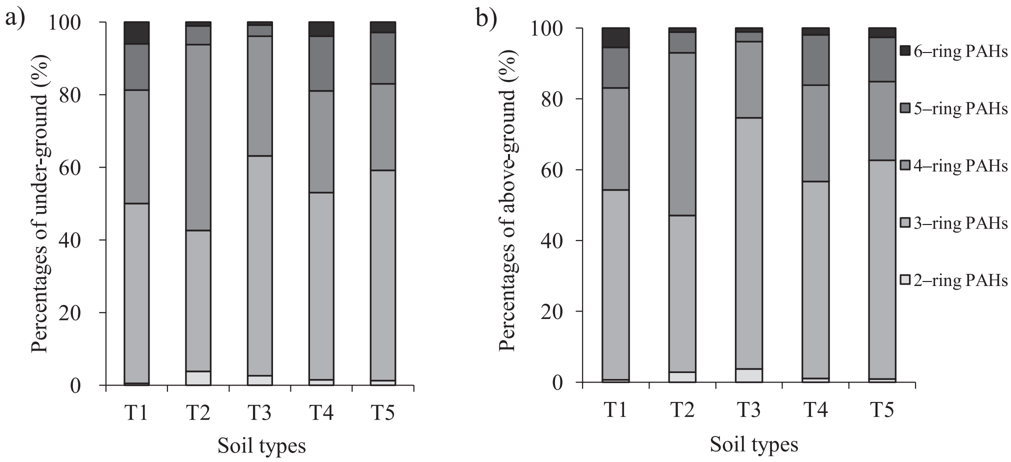

Fig. 3. Relative contributions of PAHs in radish (%): a) underground and b) aboveground.

Contribution of PAHs in Atmospherically Deposited Particulates

Σ_{16}PAHs in atmospherically deposited particles were 1,269.3 ng/g. The concentration of 4-ring PAHs was 467.6 ng/g, followed by 5- and 6-ring PAHs with concentrations of 331.6 ng/g and 279.9 ng/g, respectively (Fig. 4.). HMW-PAHs accounted for nearly 80% of total 16 PAHs in atmospherically deposited particles. Similar results have been reported in previous studies. Duan et al. [53] found that the proportion of 4-ring PAHs in the atmospheric particles of Beijing was the largest (48.7%), followed by 5-ring PAHs (32.5%) and 6-ring PAHs (14.9%). The PAHs in the atmospheric particles in spring, autumn, and winter of Huainan City were mainly 4–6-ring PAHs, accounting for 81.6%, 84.4%, and 85.6% of total PAHs, respectively [54]. Atmospheric LMW-PAHs are mainly concentrated in the gas phase, while HMW-PAHs are concentrated in the particulate phase [55-58].

Total PAH deposition amounts in the five soils were, respectively, 0.0015, 0.0005, 0.0003, 0.0014, and 0.0012 µg. The proportions of PAHs from deposited particulates to total PAHs in aboveground parts of radish planted in T1-T5 soils were 0.71%, 0.46%, 0.28%, 0.29%, and 0.13%, respectively, thereby suggesting that the atmospheric particles had a slight effect on the concentrations of PAHs in aboveground parts of radish. Fismes et al. and Khan et al. [10, 24] also showed that the PAH contribution of atmospheric particles for lettuce, which was found to be less than 1.2%, was negligible.

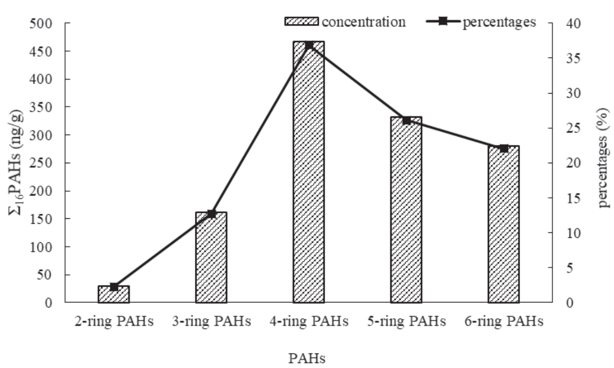

Fig. 4. Concentrations and percentages of PAHs in atmospherically deposited particulates.

Many studies have reported that PAHs in soils play an important role in PAHs in plants. PAH accumulation in vegetables increases with PAH concentrations in soil [38], suggesting that uptake of PAHs through roots is the main pathway by which PAHs can enter plants from the soil [59-60]. However, Wang [61] reported that atmospheric absorption pathway was the main way for PAHs into the vegetables. Xiong et al. [62] showed that PAH contributions from air transferred into the cabbages was higher than that of PAHs from soil near a coking production base. The composition of PAHs in plant tissues was similar to that of air in an industrial area, indicating that a gaseous path considerably contributes to PAH accumulation [13]. The relative level of PAHs in the soil and atmosphere may account for this difference. The present study was carried out in Qingdao in spring. PAH concentration in the atmosphere surrounding Qingdao during spring and summer was lower than in other seasons, and the concentration of BaP in the atmosphere was lower than that of China's national standard, thereby explaining the low concentration of PAHs in the atmosphere [63]. By comparison, soils in this study were PAH-contaminated soil, with T3 soil being heavily contaminated. This condition may explain the slight effect of atmospheric particles on PAHs in radish.

Transfer Factors of PAHs in Radish

The transfer factors of PAHs in radish planted in different soils are presented in Table 2. The values of RCFs (0.30-0.55) were significantly higher than those of SCFs (0.19-0.39, $p < 0.05$). PAH transport from the roots to the stems and leaves is difficult because PAHs cannot reach the root epidermis inside the xylem [64]. For parts of the radish found underground, the RCF of ANT and FLA was high (0.61), while the RCF value of BPE was low

Table 2. Transfer factors of PAHs in radish planted in different soils.

PAHs	RCFs		BCFs	
	Mean	S.D.	Mean	S.D.
2-ring	0.45	0.23	0.37	0.18
3-ring	0.54	0.13	0.37	0.10
4-ring	0.55	0.06	0.39	0.06
5-ring	0.37	0.07	0.22	0.06
6-ring	0.30	0.05	0.19	0.08

at 0.28. For parts of radish aboveground, the BCF of FLA was the highest (0.49). The values of RCFs and BCFs with 2–4 ring PAHs were significantly higher than 5–6-ring PAHs ($p<0.05$) due to their considerable vapor pressure, water solubility, and bioavailability. In comparison, the 5–6-ring PAHs were preferentially associated with organic matter in soil and were strongly associated with black carbon [46-48], which means that incorporating 5–6-ring PAHs into plant cells is difficult [65]. The availability and uptake rate of 5–6-ring PAHs decreases, resulting in low RCFs and BCFs [10, 24, 49].

Health Risk Assessment

Table 3 shows the TEQ_{BaP} of PAHs in the edible parts of radish planted in five soils. TEQ_{BaP} in radish ranged from 12,102.2 ng/kg to 46,621.6 ng/kg. PAH concentrations in T3 soil were the highest, but the BaP concentrations in radish of T4 and T5 soils were significantly higher than in T3 soils, and the BkF, BbF, and DBA concentrations in radish of T5 soil were significantly higher than those in other soils ($p<0.05$). The TEQ_{BaP} in radish planted in T4 (34727.6 ng/kg) and T5 (46621.6 ng/kg) soils was significantly higher than in other soils ($p<0.05$).

In this study, the ILCR of PAHs in the edible parts of radish planted in T1–T5 soils for children, adolescents, and seniors was lower than 1.0×10^{-6}. The ILCR of PAHs for adults in radish planted in T1–T5 soils were 7.8×10^{-7}, 8.0×10^{-7}, 1.1×10^{-6}, 2.2×10^{-6}, and 3.0×10^{-6}, respectively (Table 4). The ILCR values of acceptable risk level is at 1.0×10^{-6}. If the ILCR value is lower than 1.0×10^{-6}, then no risk exists. An ILCR value between 1.0×10^{-6} and 1.0×10^{-4} denotes potential risk, and ILCR values higher than 1.0×10^{-4} are markers of serious carcinogenic risk [26, 29, 66]. No carcinogenic risks through the ingestion of radish planted in T1–T5 soils exist for children, adolescents, and seniors, whereas carcinogenic risks through ingestion of radish planted in T3, T4, and T5 soils were present for adults.

Different sources of pollution in different regions indicate that PAHs in the study area had different exposures compared to those of other studies. Compared with the food exposure risk in other areas, ILCR for PAH dietary exposure through spinach, radish, cowpea, bottle gourd, bitter gourd, and ridge gourd investigated in Delhi was 3.4×10^{-6}, and the ILCR values for adults were

Table 3. TEQ_{BaP} (ng/kg) in the edible parts of radish planted in five soils.

	TEFs	T1	T2	T3	T4	T5
NAP	0.001	0.3	14.6	25.2	5.3	6.1
ACY	0.001	20.2	87.8	17.3	45.2	92.3
ACE	0.001	24.9	18.4	84.0	41.4	48.2
FLU	0.001	16.6	27.7	103.4	19.6	38.4
PHE	0.001	15.7	11.6	291.5	17.3	25.1
ANT	0.01	200.0	29.9	991.5	589.0	768.4
FLA	0.001	18.4	71.0	93.9	16.5	29.9
PYR	0.001	30.8	59.1	76.5	26.6	29.0
BaA	0.1	74.0	3,924.8	2,720.6	2,695.8	2,618.3
CHR	0.01	117.0	264.8	1,262.5	292.5	304.7
BbF	0.1	1,152.0	1,167.3	1,335.0	1,693.5	1,948.7
BkF	0.1	420.0	194.6	834.9	842.8	1,048.9
BaP	1	7,620.0	6,050.1	8,028.4	25,970.0	36,720.0
DBA	1	1,720.0	242.4	335.6	1,828.4	2,133.1
BPE	0.01	56.3	24.3	36.2	83.2	62.5
IPY	0.1	616.0	165.2	457.1	560.5	747.9
SUM		12,102.2	12,353.6	16,693.5	34,727.6	46,621.6

Table 4. ILCR values of PAHs for different age groups in radish planted in five soils.

	T1	T2	T3	T4	T5
Children	1.3×10^{-7}	1.4×10^{-7}	1.8×10^{-7}	3.8×10^{-7}	5.1×10^{-7}
Adolescents	1.1×10^{-7}	1.1×10^{-7}	1.5×10^{-7}	3.2×10^{-7}	4.2×10^{-7}
Adults	7.8×10^{-7}	8.0×10^{-7}	1.1×10^{-6}	2.2×10^{-6}	3.0×10^{-6}
Seniors	1.8×10^{-7}	1.8×10^{-7}	2.4×10^{-7}	5.0×10^{-7}	6.8×10^{-7}

4.0×10^{-5}, 4.1×10^{-5}, and 7.1×10^{-6} in Taiyuan, Lanzhou, and Linfen, respectively. These values suggest potential carcinogenic risks [26, 51, 67-68]. The preceding cities mentioned have high energy consumption and are heavily contaminated. The ILCR values of PAH dietary vegetable exposure for adults in other areas were higher than in T1 and T2 soils and close to the ILCR values in T3, T4, and T5 soils. Hence, more attention should be provided to the health risks of the vegetables growing in diesel-contaminated soils and heavily contaminated cities.

Conclusions

PAH concentration in radish was lower than in the soils. PAH concentration in parts of radish underground and aboveground had a significantly positive correlation with PAH concentration in soil. The composition of PAHs in the radish and soil were similar. The 2–4-ring PAHs were the predominant PAH compounds in radish. Buried parts of radish had higher PAH concentrations than parts aboveground in the same soil, and the RCF values of PAHs in radish were larger than BCF values. These results suggest that the root was key to the entry of PAH from the soil to the plant. Atmospherically deposited particles had a slight effect on PAH concentration in aboveground parts of radish. The total TEQ_{BaP} in radish grown in diesel-contaminated soils was higher than in other types of soils. Therefore, no carcinogenic risks to children, adolescents, and seniors exist through the ingestion of radish planted in T1–T5 soils, while carcinogenic risks were present for adults through the ingestion of radish planted in T3, T4, and T5 soils.

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