Elevated urbanization-driven plant accumulation and human intake risks of polycyclic aromatic hydrocarbons in crops of peri-urban farmlands

Anping Zhang1 · Xintao Ye2 · Xindong Yang2 · Jiacheng Li2 · Haofeng Zhu2 · Honglei Xu2 · Jiaqi Meng2 · Tianwei Xu2 · Jianqiang Sun1

Received: 17 March 2021 / Accepted: 30 April 2022 / Published online: 9 May 2022
© The Author(s), under exclusive licence to Springer-Verlag GmbH Germany, part of Springer Nature 2022

Abstract
As an ubiquitous carcinogen, polycyclic aromatic hydrocarbons (PAHs) are closely related to anthropogenic activities. The process of urbanization leads to the spatial interlacing of farmlands and urbanized zones. However, field evidence on the influence of urbanization on the accumulation of PAHs in crops of peri-urban farmlands is lacking. This study comparatively investigated the urbanization-driven levels, compositions, and sources of PAHs in 120 paired plant and soil samples collected from the Yangtze River Delta in China and their species-specific human intake risks. The concentrations of PAHs in crops and soils in the peri-urban areas were 2407.92 ng g⁻¹ and 546.64 ng g⁻¹, respectively, which are significantly higher than those in the rural areas. The PAHs in the root were highly relevant to those in the soils (R² = 0.63, p < 0.01), and the root bioconcentration factors were higher than 1.0, implying the contributions of root uptake to plant accumulations. However, the translocation factors in the peri-urban areas (1.57 ± 0.33) were higher than those in the rural areas (1.19 ± 0.14), indicating the enhanced influence through gaseous absorption. For the congeners, the 2- to 3-ring PAHs showed a higher plant accumulation potential than the 4- to 6-ring PAHs. Principal component analysis show that the PAHs in the peri-urban plants predominantly resulted from urbanization parameters, such as coal combustion, vehicle emissions, and biomass burning. The mean values of estimated dietary intake of PAHs from the consumption of peri-urban and rural crops were 9116 ng day⁻¹ and 6601.83 ng day⁻¹, respectively. The intake risks of different crops followed the order rice > cabbage > carrot > pea. Given the significant input of PAHs from urban to farmland, the influence of many anthropogenic pollutants arising from rapid urbanization should be considered when assessing the agricultural food safety.

Keywords Polycyclic aromatic hydrocarbons · Urbanization · Plant uptake · Plant accumulation · Estimated dietary intake

Introduction
With the rapid urbanization and industrialization in many developing countries, many problems, such as land insecurity, worsening of water quality, excessive air pollution, scattered waste disposal, and elevated carbon emissions, are causing environmental degradation (Song et al. 2008). The problem on various anthropogenic organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), phthalic acid esters, pesticides, polybrominated diphenyl ethers, and polychlorinated biphenyls (PCBs), has been a growing concern (Liu et al. 2014; Sun et al. 2016, 2018). Through the incomplete combustion of organic matter and the use of industrial additives, these pollutants are released, dispersed, and stored in environmental media through the pathways of airborne pollution, reclaimed water irrigation, and waste disposal (Gune et al. 2019; Witter et al. 2014; Wu et al. 2019) and consequently place tremendous direct or indirect pressure on ecosystems around cities (Karageorgou et al. 2020; Li et al. 2016). Extensive agricultural planting areas have been closely adjacent to the expanding cities from the

Responsible Editor: Elena Maestri

Jianqiang Sun
sunjianqiang@zjut.edu.cn

1 Key Laboratory of Microbial Technology for Industrial Pollution Control of Zhejiang Province, Zhejiang University of Technology, Hangzhou 310014, China

2 International Joint Research Center for Persistent Toxic Substances, College of Environment, Zhejiang University of Technology, Hangzhou 310014, China
land urbanization process, immediately interlacing urban areas and farmlands (Zhou et al. 2021). As a result of the migrations and accumulations, the risk related to the urban source environmental hazards could be magnified through agricultural production, food chains, and human intake and ultimately affect human health (Sun et al. 2021; Zhu et al. 2011). Therefore, exploring the interactive relationship between urbanization and the agricultural ecosystems around cities at the level of environmental pollutants has great scientific and practical significance.

PAH is a typical anthropogenic persistent organic pollutant with lipophilic, environmental persistent, long-distance transmitting, and toxic properties (Lou et al. 2019; White et al. 2016). PAHs are widely present in the natural environment. The US Environmental Protection Agency listed 16 PAHs as priority pollutants, and some of them are even human carcinogens (Wang et al. 2012). They are mainly produced by the incomplete combustion process and pyrolysis of organic matter. Factitious sources include traffic emissions (such as motor vehicle exhaust), industrial waste gas from coal combustion, coal emissions for commercial and household heating, and the combustion of other biomass fuels (especially the burning of straw in rural areas). PAHs are ubiquitously presented in agricultural soils and plants (Soukarieh et al. 2018; Wang et al. 2017; Waqas et al. 2014). Given that PAH pollution mainly occurs in densely populated main urban areas, industrial centers, and main traffic arteries, they can be directly transported through the atmosphere by adsorption through the surface of particles and finally deposited in soils and plants in peri-urban agricultural planting areas. For example, the average concentrations of ΣPAHs in amaranth, spinach, leeks, and rice tissue in the Yangtze River Delta of China were 1710.49, 1176.96, 1218.36, and 352.12 ng g⁻¹ dw, respectively (Wang et al. 2017).

As the main source of energy of human beings, several edible crops can absorb and transport PAHs. The primary uptake of PAHs for crops is through absorption by the root part and the aboveground tissue (Houshani et al. 2019; Wild et al. 2005; Zhang and Tao 2009). Subsequently, PAHs are bio-amplified, bio-enriched, and bio-accumulated through the food chain, causing harm to human health (Ding et al. 2013; Usman et al. 2016; Wang et al. 2012). In addition, the risk of human exposure to PAHs is objective, and the incremental lifetime cancer risk (ILCR) could be elevated through the dietary intake of contaminated edible plants. A field study proved that the concentration of PAHs in the soil in urban areas was significantly increased by urbanization (Wang et al. 2020b). Nevertheless, field evidence of the influence of urbanization on the accumulation of PAHs in plants is still lacking.

In this study, the Yangtze River Delta urban agglomeration in China was chosen as the representative area with rapid urbanization, and PAHs were chosen as the model of urbanization-driven pollutants. This study aims to elucidate the relationship between the PAHs in crops in peri-urban farmlands and the urbanization sources by collecting paired plant and soil samples from peri-urban and rural farmlands, analyzing the levels and compositions of 16 PAH congeners in different tissues, and evaluating the sources and the plant uptake and accumulations.

Materials and methods

Chemical reagents and materials

A mixture standard (containing 16 PAHs in dichloromethane, >99.0%) and 5 deuterated PAHs as surrogates (acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, perylene-d₁₂, and naphthalene-d₈, >99.0%) were purchased from Sigma-Aldrich (Shanghai, China). The physical and chemical properties of 16 PAHs are shown in Table S1.

The stock solutions of all the standards were prepared in n-hexane and stored in amber glass vials. All the organic solvents used were of HPLC grade (JT Baker, Shanghai, China). Deionized water was prepared using a Milli-Q plus water purification system (Millipore Corp, Shanghai, China). Anhydrous sodium sulfate (Na₂SO₄), silica gel (60–100 mesh), florisil (60–100 mesh), and alumina oxide (100–200 mesh) for clean the samples were baked at 400 °C for 4 h before use to eliminate potential environmental pollution.

Sampling program design

Soil and plant samples were collected from 60 locations in the Yangtze River Delta in 2019, and 50 of which are adjacent to cities. Sampling site information is provided in Table S2 and Figure S1, and a global positioning system was used to identify the precise location of each site. Soil samples (0–10 cm) were obtained at each sampling site using a handheld sampling device and collected into the corresponding container as experimental samples. Four kinds of plants were used as samples, namely, carrot (Daucus carota var. sativa Hoffm.), cabbage (Brassica pekinensis (Lour.) Rupr.), pea (Pisum sativum L.), and rice (Oryza sativa L.). Plant samples (carrot, cabbage, pea, and rice) were collected from corresponding soil sites. All collected samples were placed into a new aluminum foil bag, subsequently transported to the laboratory, and stored at −20 °C until analysis. Ten paired samples were collected from 10 rural areas for comparison with the 50 samples collected from the 50 peri-urban locations. The aboveground and the underground parts of each plant sample were analyzed separately.
Chemical treatments of samples

The sample processing method was slightly improved based on its predecessors (Cao et al. 2017; Sun et al. 2015). The samples were freeze-dried, crushed, and ground through a 100-mesh sieve, and some of the crop samples that were difficult to grind were processed using a tissue grinder. According to Soxhlet extraction, we mixed 10.0 g of soil samples with diatomaceous earth and copper powder, added 250 ng g⁻¹ of the substitute standard, and then used 160 mL of the acetone/n-hexane (1:1, v/v) mixture as the extracting solution for 48 h. Meanwhile, 20 mL of the acetone/n-hexane (1:1, v/v) mixture was used for extracting 0.2 g of crop sample ultrasonically for 20 min, and this procedure was repeated three times. Then, the extract was concentrated and purified through a silica gel column. PAHs were eluted with 40 ml of n-hexane/dichloromethane (1:1, v/v), and then the eluate was converted to hexane and concentrated to approximately 1 mL. The internal standard anthracene-d10 (>99.0%) was added before GC–MS/MS analysis. GC–MS/MS was used to detect the 16 PAHs (Table S2).

Instrumental analysis

The PAHs were measured using a SIMADAZHU GC–MS/MS-TQ8040 gas mass spectrometer with an electron impact ion source during multiple reaction monitoring (MRM). The column was an SH-Rxi-5SiLMS (30 m×0.25 mm×0.25 μm) silica capillary column. In the temperature program, the initial temperature was 60 °C and maintained for 1 min, increased to 160 °C at 10 °C/min and maintained for 1 min, increased to 180 °C at 2 °C/min and maintained for 1 min, increased to 185 °C at 0.5 °C/min and maintained for 1 min, increased to 190 °C at 1 °C/min, and finally increased to 260 °C at 2 °C/min and maintained for 5 min. The carrier gas was high purity helium with a flow rate of 1 mL/min. The inlet temperature was 250 °C. Table S3 shows the ion fragments and the retention time. Figure S2 shows the chromatogram of the PAHs.

Quality control and quality assurance

The known concentration of the tracer (250 ng) was mixed into the uncontaminated soil and crops samples for the recovery experiment. The recovery rates ranged from 84 to 118%, and a blank experiment was performed every 12 samples. The concentration with a signal-to-noise ratio of 3:1 was defined as the limit of detection (LOD), and the LOD of the PAHs was 0.016 (BKFR)–0.72 (ACY) ng/mL.

Statistical analysis

In this study, the PAH ratio and PCA were used to determine the PAH source in soil and crops. The isomer ratios of BaA/(BaA + CHR) and IcdP/(IcdP + BghiP) can be used to distinguish petroleum combustion from other types of combustion. We performed statistical analysis using the SPSS version 23.0 software and PCA to identify the possible sources of PAHs (Wang et al. 2020a). The input variables were 16 PAH concentrations measured in 50 sets of peri-urban soil and crop samples, and the three main components were extracted using PCA. We used ArcGIS 10.2 to generate the sampling maps. In addition, Origin was used to deal with the correlation between crops and soil. Bioconcentration factor (BCF) and translocation factor (TF) were calculated using the following equations:

\[
RCF = \frac{C_1}{C_2} \tag{1}
\]

\[
TF = \frac{C_3}{C_4} \tag{2}
\]

where \(C_1, C_2, C_3,\) and \(C_4\) are the concentrations of PAHs in the plants, soils, stem leaves, and roots, respectively, on a fresh weight basis (ng/g or ng/mL).

Risk assessment

The ILCR model is widely used in soil and vegetables. To assess the degree of human exposure to PAHs through the intake of crops, the following formula is used to estimate the daily intake of the local population (Li and Ma 2016; Sun et al. 2019a):

\[
CS = \Sigma Ci \times TEFi \tag{3}
\]

\[
EDI = CS \times IR \tag{4}
\]

\[
ILCR = \frac{CSF \times EDI \times EF \times ED \times CF}{BW \times AT} \tag{5}
\]

where the EDI is the estimated daily intake through the consumption of crops/soils (ng/d) and ILCR is the increase in the lifetime cancer risk of an individual. As shown in Table S4 in Supplementary Material, CSF represents the carcinogenic slope factor of ingestion, that is, 7.3 (mg/kg/day)⁻¹ (USEPA 1991). IR is the intake rate (mg day⁻¹), and the IR values of cabbage, carrot, pea, and rice are 147670, 71950, 3780, and 252690, respectively (Duan 2013). EF is the exposure frequency (day/year), ED is the duration of PAH exposure (years), BW is the body weight (kg), AT is the average life span (days), and CS is the sum of the specific PAH concentrations calculated by the toxicity equivalence factor (TEF) method. Ci is the concentration of...
each PAH (mg/kg), and TEFi is the corresponding toxicity equivalent coefficient (Table S1). CF is the conversion factor ($10^{-6}$ mg/ng).

**Result and discussion**

**Elevated PAH concentrations in plant**

All the plant samples were divided into aboveground tissue (leaf or fruit) and underground tissue (root). The total concentrations of PAHs in the plants ranged from 984.88 to 3888.46 ng g$^{-1}$ with an average of 2192.43 ng g$^{-1}$.

As shown in Fig. 1, the PAH concentration of the aboveground part was significantly higher than that of the root part ($p < 0.01$). The average concentrations of PAHs in the aboveground and root tissues were 2645.6 and 1846.8 ng g$^{-1}$, respectively. In both tissues of all the species, the average concentrations of PAHs in the peri-urban locations were higher than those in the rural areas. Among the different kinds of plants, the average concentrations in the entire plants followed the order carrot > cabbage > pea > rice. However, when the edible parts of each species were compared, the order changed to cabbage > pea > rice > carrot. The PAH concentrations in the aboveground tissue among the four species have a large difference, while those in root are relatively close (Fig. 1 and Table 1). The PAH concentration in the edible parts of leafy vegetables is higher than that in the root-edible plant, which may be affected by two factors, namely, the absorption and transport of soil roots and the absorption of gaseous pollutants by leaves (Li et al. 2020). Plant accumulation is affected by its own absorption capacity and metabolic potential and the intensity of exogenous pollution (Sun et al. 2015, 2021). Therefore, the relationship between the aboveground and underground parts must be established, and the soil pollution, the composition, and the source trace must be analyzed further.

**Concentrations of PAHs in agricultural soil**

The total concentrations of PAHs in the 60 groups of soil samples range from 109.5 to 2451.89 ng g$^{-1}$, with an average of 507 ng g$^{-1}$. The concentrations of PAHs in soil presented in this paper are of medium magnitude and comparable to those in the Yangtze River Delta Urban agglomeration of China (Wang et al. 2017), Shanghai, China (Yang et al. 2020), and Khyber Pakhtunkhwa Province, Pakistan (Waqas et al. 2014) and lower than those determined in Lebanon (Soukarieh et al. 2018) and the Dilovasi in Kocaeli region of Turkey (Cetin 2016).

**Table 1** Concentration of PAHs in different crops

| Plant tissue      | Species | LMW PAHs (ng/g) | Mean | Median | Min | Max | HMW PAHs (ng/g) | Mean | Median | Min | Max | PAHs (ng/g) | Mean |
|-------------------|---------|-----------------|------|--------|-----|-----|-----------------|------|--------|-----|-----|------------|------|
| Aboveground part  | Cabbage | 2189            | 1980.71 | 906.95 | 4288.90 | 551.50 | 531.97 | 159.78 | 1163.85 | 2741.49 |
|                   | Carrot  | 2412.56         | 2669.48 | 1249.98 | 2949.18 | 1068.73 | 563.24 | 331.73 | 3692.21 | 3558.65 |
|                   | Pea     | 1779.72         | 1890.66 | 1230.55 | 2178.17 | 643.72 | 504.05 | 390.45 | 1221.95 | 2423.44 |
|                   | Rice    | 1640.79         | 1533.90 | 821.54 | 2930.87 | 474.22 | 452.39 | 117.52 | 691.29 | 2116.77 |
| Underground part  | Cabbage | 1355.70         | 1269.47 | 729.02 | 2554.51 | 518.77 | 467.35 | 187.24 | 1289.52 | 1874.18 |
|                   | Carrot  | 1489.44         | 1411.66 | 744.47 | 2301.33 | 494.99 | 392.40 | 92.62 | 1295.73 | 2048.94 |
|                   | Pea     | 1150.30         | 1079.22 | 860.94 | 1579.85 | 483.94 | 507.12 | 292.62 | 719.12 | 1635.79 |
|                   | Rice    | 1285.37         | 1232.65 | 672.78 | 1750.01 | 498.92 | 460.39 | 219.46 | 1156.83 | 1784.32 |

Fig. 1 Concentrations of PAHs in crops. (a) Peri-urban and rural crops; (b) different crops
The total amount of PAHs in the farmlands in the 50 peri-urban areas was 547.42 ng g⁻¹, and that in the 10 suburban areas was 310.57 ng g⁻¹. Both plants and soil are higher in peri-urban than in rural areas. As a city originated pollutant, similar trends were world-widely observed in many countries. For instance, in Lebanon, the BaPeq values in industrial and urban soils were 777 and 256 times higher than those in rural soil, respectively (Soukarieh et al. 2018). The concentration of PAHs in the urban areas of the Monterrey Metropolitan Area, Mexico, is higher than that in the rural areas (Lopez-Ayala et al. 2019). Therefore, the process of urbanization has a certain impact on the emission of PAHs, which is also the reason why the concentration of PAHs in urban crops is higher than that in the rural areas.

**Plant uptake of PAHs from agricultural soil**

Soil and plant are a pair of indispensable components in the agricultural environment. The fate of PAHs in soil–plant systems could be attributed to their transfer process by connecting its levels in soil and plant. Figure 2 shows that a good correlation exists between the root part concentrations and the soil concentrations ($R^2=0.63$, $p<0.01$), indicating that PAHs migrate from farmland soil to the roots of plants through root uptake (Tao et al. 2009; Wei et al. 2021; Wild et al. 2005). However, the correlation between the aboveground part and soil is not high as that between the root part and soil ($R^2=0.31$, $p<0.01$). This phenomenon might be the result of different environmental factors, including traffic source and biomass combustion, which lead to differences in the adsorption process through leafy uptake (Zhang et al. 2019). The absorption of PAHs in the gas phase reportedly accounts for 90.6% of the total absorption of vegetables, and the absorption of PAHs in soil accounts for 9.4% of the total absorption of vegetables (Jia et al. 2019). Therefore, the urbanization process has an impact on the pollution of the aboveground tissue.

All the bioconcentration factors (BCFs) of the root exceed 1.0, implying the contributions of root uptake to the plant accumulations. No significant difference in BCF value exists between different species. However, the concentrations in the aboveground tissue are higher than those in the root, probably because the aboveground part is more prone to accumulate PAHs than the root part. The translocation factors (TFs) in the peri-urban areas ($1.57\pm0.33$) are higher than those in the rural area ($1.19\pm0.14$), indicating the enhanced influence through gaseous absorption. In these samples, the aboveground tissue of low-molecular-weight (LMW, < 4 rings) accounts for 41–93%, with an average of 78%, and the underground tissue of LMW accounts for 55–90%, with an average of 73%, indicating that LMW has a greater contribution to the aboveground part than to the underground part. The TFs of the LMW PAHs also exceed 1.0, indicating their easy accumulation in crops, which is consistent with the results of previous studies (Ding et al. 2013, Jia et al. 2018, Waqas et al. 2014) (Figs. S3–S6). This phenomenon may be attributed to the high water solubility and vapor pressure of LMW PAHs, which facilitate their absorption by the root and aboveground parts (Li et al. 2001).

By contrast, high-molecular-weight (HMW, < 4 rings) PAHs can easily accumulate in the soil compared with LMW PAHs (Huang et al. 2017). This phenomenon may be attributed to the disappearance of LMW PAHs through photolysis and volatilization. This result is consistent with those of previous studies (Marques et al. 2016a, b; Qu et al. 2020). In urban areas with high pollution levels, soil may become the ultimate environmental fate of PAHs through atmospheric deposition (Agarwal et al. 2009; Feng et al. 2017; Zheng et al. 2015). Some studies point out that the HMW PAHs in soil easily enter the human body through sebum (Beriro et al. 2020). Therefore, the HMW PAHs in soil pose a higher
risk to human health than the LMW PAHs because of their higher concentrations.

**Sources related with urbanization**

The PAH isomer ratio has been widely used in environmental media for source identification. In the soil samples, the BaA/(BaA + CHR) ratios exceed 0.35, and the IcdP/(IcdP + BghiP) ratios exceed 0.5, indicating that the combustion of biomass and coal is the main source of PAHs in soil. For the crop samples, the results are consistent with those of the soil samples (Figs. 3 and S7), which is consistent with the conclusion of Wang et al. in their study of the Yangtze River Delta (Wang et al. 2017).

To explore the composition and sources of PAHs in peri-urban soils and crops, PCA is used to obtain the component contributions from different sources. The aboveground and root parts of crops and soil have three main components, which account for more than 70% of the total components (Tables S5–S7). PC1 in soil accounts for 50.267%, of which FRT (0.865), PYR (0.722), BaAN (0.935), CHR (0.948), BbFR (0.939), BkFR (0.821), BaP (0.947), IcdP (0.853), DahA (0.823), and BghiP (0.876) account for a large proportion, reflecting the emissions from transportation and industry exhaust and the combustion of petroleum (Sari et al. 2020). These are some elements related to the city. PC2 accounts for 16.414%, of which ACY (0.544), PHE (0.820), and ANT (0.865) account for a large proportion. PC2 indicates the combustion of biomass and coal. PC3 accounts for 9.403%, of which NAP (0.341), FLR (0.685), and ACE (0.630) account for the most part. PC3 represents the mixture of crude oil emissions and coal combustion (Tables S5–S7) (Qu et al. 2020).

For both the aboveground and underground tissues of peri-urban crops, PC1, which is dominated by FLR, PYR, BaAN, CHR, BbFR, BkFR, BaP, IcdP, DahA, and BghiP, reflects the traffic and industrial waste gas emissions and the burning of oil and coal. ACE, FRT, and PHE dominate the second component (PC2), indicating that the combustion of biomass and coal is the main source. NAP, ACY, and ANT mainly indicate that the third component (PC3) is the emissions from crude oil and the combustion of diesel. Therefore, unlike the relatively simple source in rural areas, the PAHs pollution in cities mainly comes from traffic emissions, followed by biomass and petroleum combustion.

**Health risk assessment**

The ingestion of vegetables and soil contact are major PAH exposure pathways for humans. Thus, we are concerned about crops and soil. In this study, the ILCR and the EDI were calculated based on ingestion to assess the potential health risks imposed by the PAHs in soil and crops to humans in different age groups (Sun et al. 2019b). According to the literature, we conducted a comparative analysis between urban and rural areas, different plants, and adults and children (Table 2) (Wang et al. 2018). Generally, an ILCR lower than $10^{-6}$ is considered safe, an ILCR between $10^{-6}$ and $10^{-4}$ indicates low risk, and an ILCR above $10^{-4}$ indicates health problems.

In soil, the ILCR result indicates that almost all samples were contaminated with PAHs below the acceptable risk level, in which are consistent with previous studies (Zhou et al. 2021). The edible part of the vegetable is not the same as the soil, which the ILCR and EDI values are higher in cities than in rural areas. This may be caused by more serious levels of pollution in the city.

Table 2 Human ingestion risk of PAH of edible parts

|                  | ILCR               | EDI (ng/d) |
|------------------|--------------------|------------|
| Adults           | Children           |            |
| Peri-urban       | 2.06712E−04        | 3.23468E−04| 9116.00    |
| Rural            | 1.49701E−04        | 2.35779E−04| 6601.83    |
| Cabbagge         | 1.98775E−04        | 3.09679E−04| 8765.99    |
| Carrot           | 1.34855E−04        | 2.12397E−04| 5947.12    |
| Pea              | 1.41559E−05        | 2.22956E−05| 624.28     |
| Rice             | 3.44266E−04        | 5.4222E−04 | 15182.15   |

![Fig. 3 Isomer ratio and source analysis of PAHs of aboveground part (a) and underground part (b) in peri-urban crops](image-url)
Therefore, we further analyze the risk involved in the ingestion of crops from the peri-urban areas. For adults and children, 84% of the samples have an ILCR risk higher than \(10^{-6}\), and the ILCR of the children is slightly higher than that of the adults, indicating that the exposure risk of children is higher than that of adults. The more high-risk period for crop and soil PAH contamination is childhood (Wang et al. 2017). Among different crops, peas have the lowest risk, which is less than \(10^{-6}\). Those of the other crops exceed \(10^{-4}\), which indicate health risks. The intake risks of different crops follow the order rice > cabbage > carrot > pea. Rice has the highest risk because of its relatively high daily intake, followed by cabbage because of its large leaves and high pollutant concentration (Table 2). Rice and cabbage are the most easily accessible crops, so their intake risks deserve attention. Thus, the long-term cumulative risk of the intake of these crops to the human body cannot be ignored.

**Conclusion**

This study explored the levels, compositions, and sources of PAHs in soil and crops in the peri-urban farmlands in the Yangtze River Delta urban agglomeration of China and the influence of urbanization on their plant accumulation and human intake risks. The concentrations of PAHs in soil and crops in the peri-urban farmlands are generally higher than those in the rural areas, indicating the relatively higher risk of ingestion of those in the agricultural products planted in the peri-urban zones. A significant correlation between the PAHs in the plant roots and the corresponding soil samples was observed, and the calculated root BCF values exceed 1.0, implying the contributions of the root uptake from the urbanization-influenced PAHs in soils to the elevated plant accumulations. However, the correlation between the above-ground part and soil was not as high as that between the root part and soil. Furthermore, the concentrations in the above-ground part were generally higher than those in the root part, with an average TF value of 1.57 ± 0.33 in the peri-urban areas, indicating the enhanced influence through the pathway of gaseous absorption of urban source PAHs. In addition, the main sources of PAHs in crops are anthropogenic activities, such as traffic emissions and the combustion of coal and oil. Thus, under the influence of cities, the plant uptake and accumulation of PAHs in peri-urban farmlands could be elevated through two pathways, namely, root uptake from soils contaminated by urban-source-PAHs and direct gaseous uptake through plant leaves. This study has a certain reference significance for alleviating the adverse effects of urbanization and reducing the impact of urban anthropogenic activities on agricultural ecosystems, such as conservation of farmlands in urban planning steps and planting low-ingestion species of crops.

**Supplementary Information** The online version contains supplementary material available at https://doi.org/10.1007/s11356-022-20623-1.

**Author contribution** Anping Zhang: Conceptualization and writing—review and editing. Xintao Ye: Writing (original draft) and formal analysis. Xindong Yang: Methodology and software. Jiacheng Li: Methodology. Haoeng Zhu: Investigation. Honglei Xu: Validation. Jiaqi Meng: Visualization. Tianwei Xu: Investigation. Jianqiang Sun: Writing (review and editing) and supervision.

**Funding** This study was supported by the National Natural Science Foundation of China (21976160, 21577127).

**Data availability** The authors declare that (the/all other) data supporting the findings of this study are available within the article (and its supplementary material files).

**Declarations**

**Ethics approval and consent to participate** This study does not report on or involve the use of any animal or human data or tissue.

**Consent for publication** The authors agree to publication in the *Journal of Environmental Science and Pollution Research* and also to publication of the article in English by Springer in Springer’s corresponding English-language journal.

**Competing interests** The authors declare no competing interests.

**References**

Agarwal T, Khililar PS, Shridhar V, Ray S (2009) Pattern, sources and toxic potential of PAHs in the agricultural soils of Delhi. India J Hazard Mater 163(2–3):1033–1039

Beriro DJ, Cave M, Kim A, Craggs J, Wragg J, Thomas R, Taylor C, Nathaniel CP and Vane C (2020) Soil-sebum partition coefficients for high molecular weight polycyclic aromatic hydrocarbons (HMW-PAH). J Hazard Mater 398

Cao H, Chao S, Qiao L, Jiang Y, Zeng X, Fan X (2017) Urbanization-related changes in soil PAHs and potential health risks of emission sources in a township in Southern Jiangsu, China. Sci Total Environ 575:692–700

Cetin B (2016) Investigation of PAHs, PCBs and PCNs in soils around a heavily industrialized area in Kocaeli, Turkey: concentrations, distributions, sources and toxicological effects. Sci Total Environ 560–561:160–169

Ding C, Ni HG, Zeng H (2013) Human exposure to parent and halogenated polycyclic aromatic hydrocarbons via food consumption in Shenzhen, China. Sci Total Environ 443:857–863

Duan X (2013) Exposure factors handbook of Chinese population (adults). China Environment Science Press, Beijing

Feng DL, Liu Y, Gao Y, Zhou JX, Zheng LR, Qiao G, Ma LM, Lin ZF, Grathwohl P (2017) Atmospheric bulk deposition of polycyclic aromatic hydrocarbons in Shanghai: temporal and spatial variation, and global comparison. Environ Pollut 230:639–647

Gune MM, Ma WL, Sampath S, Li WL, Li YF, Udayashankar HN, Balakrishna K, Zhang ZF (2019) Occurrence of polycyclic aromatic hydrocarbons (PAHs) in air and soil surrounding a coal-fired thermal power plant in the south-west coast of India. Environ Sci Pollut Res 26(22):22772–22782

Houshani M, Salehi-Lisar SY, Motakakerazad R, Movafeghi A (2019) Uptake and distribution of phenanthrene and pyrene in...
roots and shoots of maize (Zea mays L.). Environ Sci Pollut Res 26(10):9938–9944
Huang YP, Liu M, Wang RQ, Khan SK, Gao DZ, Zhang YZ (2017) Characterization and source apportionment of PAHs from a highly urbanized river sediments based on land use analysis. Chemosphere 184:1334–1345
Jia JP, Bi CJ, Zhang JF, Jin XP, Chen ZL (2018) Characterization of polycyclic aromatic hydrocarbons (PAHs) in vegetables near industrial areas of Shanghai, China: sources, exposure, and cancer risk. Environ Pollut 241:750–758
Jia JP, Bi CJ, Zhang JF, Chen ZL (2019) Atmospheric deposition and vegetable uptake of polycyclic aromatic hydrocarbons (PAHs) based on experimental and computational simulations. Atmos Environ 204:135–141
Kameda T (2011) Atmospheric chemistry of polycyclic aromatic hydrocarbons and related compounds. J Health Sci 57(6):504–511
Karageorgou K, Manoli E, Kouras A, Samara C (2020) Commuter exposure to particle-bound polycyclic aromatic hydrocarbons in Thessaloniki, Greece. Environ Sci Pollut Res 28:59119–59130
Li H, Ma Y (2016) Field study on the uptake, accumulation, translocation and risk assessment of PAHs in a soil–wheat system with amendments of sewage sludge. Sci Total Environ 560–561:55–61
Li GD, Fang CL, Wang SJ, Sun S (2016) The effect of economic growth, urbanization, and industrialization on fine particulate matter (PM2.5) concentrations in China. Environ Sci Technol 50(21):11452–11459
Li Y, Liu M, Li RK, Sun P, Xia HB and He TH (2020) Polycyclic aromatic hydrocarbons in the soils of the Yangtze River Delta. Urban Agglomeration, China: influence of land cover types and urbanization. Sci Total Environ 715
Liu HH, Hu YJ, Luo P, Bao LJ, Qiu JW, Leung KM, Zeng EY (2014) Occurrence of halogenated flame retardants in sediment off an urbanized coastal zone: association with urbanization and industrialization. Environ Sci Technol 48(15):8465–8473
Lopez-Ayala O, Gonzalez-Hernandez LT, Alcantar-Rosales VM, Elizarraraz-de la Rosa D, Heras Ramirez ME, Silva-Vidaurri LG, Alfaro-Barbosa JM, Gaspar-Ramirez O (2019) Levels of polycyclic aromatic hydrocarbons associated with particulate matter in a highly urbanized and industrialized region in northeastern Mexico. Atmos Pollut Res 10(5):1655–1662
Lou XY, Wu PR, Guo Y (2019) Urinary metabolites of polycyclic aromatic hydrocarbons in pregnant women and their association with a biomarker of oxidative stress. Environ Sci Pollut Res 26(26):27281–27290
Marques M, Mari M, Audi-Miro C, Sierra J, Soler A, Nadal M, Domingo JL (2016a) Climate change impact on the PAH photodegradation in soils: characterization and metabolites identification. Environ Int 89–90:155–165
Marques M, Mari M, Audi-Miro C, Sierra J, Soler A, Nadal M, Domingo JL (2016b) Photodegradation of polycyclic aromatic hydrocarbons in soils under a climate change base scenario. Chemosphere 148:495–503
Park JS, Wade TL, Sweet S (2001) Atmospheric distribution of polycyclic aromatic hydrocarbons and deposition to Galveston Bay, Texas, USA. Atmos Env 35(19):3241–3249
Qu Y, Gong Y, Ma J, Wei H, Liu Q, Liu L, Wu H, Yang S, Chen Y (2020) Potential sources, influencing factors, and health risks of polycyclic aromatic hydrocarbons (PAHs) in the surface soil of urban parks in Beijing. China Environ Pollut 260:114016
Sari MF, Esen F and Tasdemir Y (2020) Characterization, source apportionment, air/plant partitioning and cancer risk assessment of atmospheric PAHs measured with tree components and passive air sampler. Environ Res 110508
Song JH, Webb A, Parmenter B, Allen DT, McDonald-Buller E (2008) The impacts of urbanization on emissions and air quality: comparison of four visions of Austin, Texas. Environ Sci Technol 42(19):7294–7300
Soukarieh B, El Hawari K, El Hussein M, Budzinski H, Jaber F (2018) Impact of Lebanese practices in industry, agriculture and urbanization on soil toxicity. Evaluation of the polycyclic aromatic hydrocarbons (PAHs) levels in soil. Chemosphere 210:85–92
Sun J, Wu X, Gan J (2015) Uptake and metabolism of phthalate esters by edible plants. Environ Sci Technol 49(14):8471–8478
Sun J, Wang Q, Zhuang S, Zhang A (2016) Occurrence of polybrominated diphenyl ethers in indoor air and dust in Hangzhou, China: level, role of electric appliances, and human exposure. Environ Pollut 218:942–949
Sun J, Xu Y, Zhou H, Zhang A, Qi H (2018) Levels, occurrence and human exposure to novel brominated flame retardants (NBFRs) and dechlorane plus (DP) in dust from different indoor environments in Hangzhou, China. Sci Total Environ 631–632:1212–1220
Sun J, Wu Y, Jiang P, Zheng L, Zhang A, Qi H (2019a) Concentration, uptake and human dietary intake of novel brominated flame retardants in greenhouse and conventional vegetables. Environ Int 123:436–443
Sun J, Yang X, Shen H, Xu Y, Zhang A, Gan J (2021) Uptake and metabolism of nonylphenol in plants: isomer selectivity involved with direct conjugation. Environ Pollut 279:116064
Tao YQ, Zhang SZ, Zhu YG, Christie P (2009) Uptake and acropetal translocation of polycyclic aromatic hydrocarbons by wheat (Triticum aestivum L.) grown in field-contaminated soil. Environ Sci Technol 43(10):3556–3560
USEPA (1991) Risk assessment guidance for superfund, Volume1, Human health evaluation manual (Part B, Development of Risk-based Preliminary Remediation Goals), EPA/540/R-92/003 Publication 9285.7–01B. http://nepis.epa.gov/Exe/ZyPURL.cgi? Dockeye=100020L.F.txt
Usman M, Hanna K, Hadarlein S (2016) Fenton oxidation to remediate PAHs in contaminated soils: a critical review of major limitations and counter-strategies. Sci Total Environ 569:179–190
Wang Y, Tian Z, Zhu H, Cheng Z, Kang M, Luo C, Li J, Zhang G (2012) Polycyclic aromatic hydrocarbons (PAHs) in soils and vegetation near an e-waste recycling site in South China: concentration, distribution, source, and risk assessment. Sci Total Environ 439:187–193
Wang J, Zhang XF, Ling WT, Liu R, Liu J, Kang FX, Gao YZ (2017) Contamination and health risk assessment of PAHs in soils and crops in industrial areas of the Yangtze River Delta region, China. Chemosphere 168:976–987
Wang CH, Zhou SL, Song J, Wu SH (2018) Human health risks of polycyclic aromatic hydrocarbons in the urban soils of Nanjing, China. Sci Total Environ 612:750–757
Wang P, Mi WY, Xie ZY, Tang JH, Apel C, Joerss H, Ebinghaus R and Zhang QH (2020a) Overall comparison and source identification of PAHs in the sediments of European Baltic and North Seas, Chinese Bohai and Yellow Seas. Sci Total Environ 737
Wang Y, Bao MJ, Zhang YW, Tan F, Zhao HX, Zhang QN and Li QL (2020b) Polycyclic aromatic hydrocarbons in the atmosphere and soils of Dalian, China: source, urban-rural gradient, and air-soil exchange. Chemosphere 244
Waya S, Khan S, Chao C, Shamshad I, Qamar Z, Khan K (2014) Quantification of PAHs and health risk via ingestion of vegetables in Khyber Pakhtunkhwa Province, Pakistan. Sci Total Environ 497:448–458
Wei BK, Liu C, Bao JS, Wang Y, Hu JC, Qi M, Jin J, Wei YJ (2021) Uptake and distributions of polycyclic aromatic hydrocarbons
in cultivated plants around an E-waste disposal site in Southern China. Environ Sci Pollut Res 28(3):2696–2706
White AJ, Bradshaw PT, Herring AH, Teitelbaum SL, Beyea J, Stellman SD, Steck SE, Mordukhovich I, Eng SM, Engel LS, Conway K, Hatch M, Neugut AI, Santella RM, Gammon MD (2016) Environ Int 89–90:185–192
Wild E, Dent J, Thomas GO, Jones KC (2005) Direct observation of organic contaminant uptake, storage, and metabolism within plant roots. Environ Sci Technol 39(10):3695–3702
Witter AE, Nguyen MH, Baidar S, Sak PB (2014) Coal-tar-based seal-coated pavement: a major PAH source to urban stream sediments. Environ Pollut 185:59–68
Wu Y, Sun J, Zheng C, Zhang X, Zhang A, Qi H (2019) Phthalate pollution driven by the industrial plastics market: a case study of the plastic market in Yuyao City, China. Environ Sci Pollut Res Int 26(11):11224–11233
Yang J, Sun P, Zhang X, Wei XY, Huang YP, Du WN, Qadeer A, Liu M, Huang Y (2020) Source apportionment of PAHs in roadside agricultural soils of a megacity using positive matrix factorization receptor model and compound-specific carbon isotope analysis. J Hazard Mater 403:123592
Zhang YX, Tao S (2009) Global atmospheric emission inventory of polycyclic aromatic hydrocarbons (PAHs) for 2004. Atmos Environ 43(4):812–819
Zhang J, Li R, Zhang X, Bai Y, Cao P, Hua P (2019) Vehicular contribution of PAHs in size dependent road dust: a source apportionment by PCA-MLR, PMF, and Unmix receptor models. Sci Total Environ 649:1314–1322
Zheng Q, Nizzetto L, Liu X, Borga K, Starrfelt J, Li J, Jiang YS, Liu X, Jones KC, Zhang G (2015) Elevated mobility of persistent organic pollutants in the soil of a tropical rainforest. Environ Sci Technol 49(7):4302–4309
Zhou B-B, Aggarwal R, Wu J, Lv L (2021) Urbanization-associated farmland loss: a macro-micro comparative study in China. Land Use Policy 101:105228
Zhu YG, Ioannidis JP, Li H, Jones KC, Martin FL (2011) Understanding and harnessing the health effects of rapid urbanization in China. Environ Sci Technol 45(12):5099–5104

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.