

Original Research

Polycyclic Aromatic Hydrocarbons in Home-Grown Allium Fistulosum from an Industrial City of China: Concentrations, Source and Cancer Risks

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Received: 27 December 2021

Accepted: 3 May 2022

Abstract

Polycyclic aromatic hydrocarbons (PAHs) in vegetables may pose a cancer risk via consumption of contaminated vegetables. Thus, this study aims to analyze the concentrations of PAHs in home-grown allium fistulosum from an industrial city of China, and assess the exposure risks. The results indicate that adsorption of PAHs through roots and local anthropogenic pollution were major factors affecting accumulation of PAHs in edible parts of allium fistulosum. The sources of PAHs were identified as vehicle emissions, agricultural waste burning and pollution discharges of petroleum factories. Daily dietary exposures of consumers to PAHs in allium fistulosum ranged from 2.76×10^{-4} to 7.02×10^{-5} ng/(d·kg), children had the highest daily exposure dose, while the teenagers had the lowest. For individuals of different gender and age groups (children, teenagers, adults and old), the incremental lifetime cancer risks (ILCRs) ranged from 1.4×10^{-6} to 3.5×10^{-5} , which were all higher than safe risk level of 10^{-6} . The findings of this study suggest that growing allium fistulosum in the urban areas of an industrial city may pose low cancer risks to consumers.

Keywords: polycyclic aromatic hydrocarbons, allium fistulosum, dietary exposure, cancer risk assessment

Introduction

The health threats posed by polycyclic aromatic hydrocarbons (PAHs) have drawn increasing concerns due to their potential for carcinogenicity, teratogenicity

and mutagenicity, especially 16 PAHs have been listed as priority pollutants by USEPA [1-3]. PAHs are ubiquitously distributed in the environment, such as soil, water, sediment and air [4, 5], and can be accumulated in the vegetables due to their lipophilicity and hydrophobicity [6, 7]. In China, about 36 million hectares farmland are threatening the organic pollution, such as PAHs [8]. The consumption of vegetables

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is an important PAHs exposure route, and accounted for >70% of total exposure in non-smoking individuals [9].

Therefore, the exposure route of PAHs via vegetable consumption should be considered for human health risk assessment.

The sources of PAHs are primarily derived from industrial discharges and incomplete combustion of fossil fuels and biomass, such as coal, gasoline, diesel and wood [10-12]. The PAHs pollution in soils and air of industrial areas are expected to be seriously, and the vegetables grown near industrial areas may be contaminated with PAHs [13]. PAHs concentrations in soils have a great influence on the concentrations in vegetables, previous study indicates that the concentrations in vegetables increased with the increase of PAHs concentrations in soils [14]. In addition, atmospheric adsorption has been regarded as another major source for PAHs in vegetables, especially for above ground parts of leafy vegetables [15], and transfer of PAHs from air to leaf is determined by gas-leaf partitioning and trapped deposited particles (wet and dry) [16].

Jilin city is an industrial city, many local residents grow *allium fistulosum* at yards for their own consumption. However, no studies on this exposure risks have been reported. In this study, we examined the concentrations of PAHs in home-grown *allium fistulosum* from Jilin city, China. In addition, a cancer risk of consumption of leafy vegetables could estimate the safety of exposure to organic pollutants in leafy vegetables [17]. The objectives of this study are to: analyze the concentrations of 16 PAHs in home-grown *allium fistulosum* from urban areas of Jilin city; explore spatial distribution of PAHs in *allium fistulosum*; identify the major sources of PAHs in *allium fistulosum*; and assess the exposure and cancer risks of PAHs via the consumption of *allium fistulosum*. This study concerns the security of vegetables grown in the urban areas near industrial areas, and gives a suggestion on consumption of vegetables grown near industrial areas.

Materials and Methods

The Study Area and Sample Collection

The study area - Jilin city is located in Jilin province of China (Fig. 1), where the average annual temperature is 3-5°C with 120 frost-free days, annual precipitation is 700 mm, and the major soil property is black soil. Jilin city is an industrial city, and has the largest deep-processing petroleum industry in Jilin province.

A total of 27 home-grown *allium fistulosum* samples were collected from nine residential plots (Fig. 1), every three samples were collected from each residential plot, and the concentrations of PAHs in one residential plots were the average concentrations of PAHs in three samples. The major PAHs pollution source is located in north of Jilin city, China. Four sampling sites (HG, SQ, TT and JH) are located around the petroleum industry, two sampling sites (TT and SJ) are located in the other side from deep-processing petroleum industry, and three (ZJ, WX and YL) are located in the south of Jilin city.

The *allium fistulosum* were grown in the middle of September 2019, and harvested in May 2020. The *allium fistulosum* samples were washed with tap water and risen with deionized water, then the cleaned *allium fistulosum* samples were partitioned into roots and edible parts. The treated *allium fistulosum* samples were air-dried in the dark, ground and passed through a 100-mesh sieve.

Chemical Analysis

Sample extraction: The extraction method was adopted from a published article [18]. The samples (10 g of *allium fistulosum* samples (dry weight)) were mixed with anhydrous sodium sulfate (1:1/w:w), added n-hexane/dichloromethane mixture (1:1 v:v), and extracted in an ultrasonic bath for 20 min (repeated 3 times). The extracts were concentrated to 1 mL

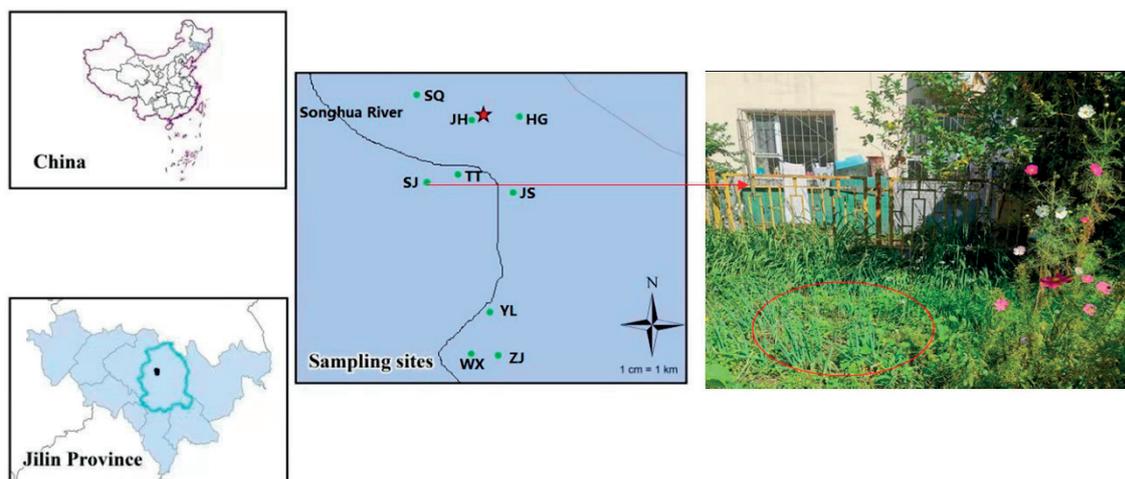


Fig. 1. The locations of study area and sampling sites. Asterisk: petrochemical industrial corporation.

and passed through silica/anhydrous sodium sulfate cartridge Cleanert Florisil 1000 mg/6 mL cartridge, Agela echnologies Inc., the USA). The cartridges were cleaned with 10 mL of n-hexane, eluted with 30 mL of n-hexane/dichloromethane mixtures (1:1 v/v), and concentrated to 1 mL for GC-MS analysis.

Gas Chromatography-Mass Spectrometry analysis: Sixteen PAHs in allium fistulosum were analyzed by an Agilent 7890B GC equipped with an Agilent 5977B mass spectrometer. The selected ion monitoring mode (SIM) was employed.

Gas Chromatography: the temperature of injection was 2800°C, HP-5 (30m×0.25mm) was applied, the injection volume was 1µL, carrier gas was helium (purity: 99.99%, rate: 1.0 mL/min). Temperature programming: initial, 80°C (maintain 2 min); increase 180°C to at a rate of 20°C/min (maintain 5 min); raise to 290°C at a rate of 10°C/min (maintain 5 min).

Mass Spectrometry: electron impact ionization and selected ion monitoring mode were employed, ionization energy was set to 70 eV, interface temperature was 280°C, the temperature of ion source was 230°C, quadrupoles temperature was 150°C, mass scanning range was 45-450 u, solvent delay time was 5 min.

Quality Control and Assurance

The method of conducting recoveries was adopted from previous study [19]. Recoveries were checked by spiking deuterated-PAHs (naphthalene-d8, acenaphthene-d10, phenantrene-d10, chrysene-d12, and perylene-d12) onto the samples. Blank and parallel samples were regularly analyzed every six samples. The target compounds were undetected in the blank samples. The recoveries of deuterated-PAHs were 75.4% to 86.2%. The method detection limits (MDL) were 0.02 to 0.17 ng/g.

Estimation of Dietary Exposure and Cancer Risk

The total B[a]P equivalent concentration is calculated using Equation (1) [20]:

$$\text{BEC} = (\text{C}_1 + \text{C}_2 + \dots + \text{C}_n) \times \text{TEF}_i \quad (1)$$

where BEC is the total B[a]P equivalent concentration in the edible part of allium fistulosum (ng/g dw); C is the monitoring concentration of PAHs in the edible part of allium fistulosum (ng/g dw); TEF_i is the corresponding toxic equivalence factor, the values of TEF are represented in Table 1.

The daily dietary exposure dose (ED) for each group (children (4-10 years); teen-agers (11-17 years); adults (18-60 years); old (>60 years) is calculated using Equation (2) [21]:

$$\text{ED} = [\text{BEC} \times \text{IR} \times \text{CF}]/\text{BW} \quad (2)$$

Table 1. The factors of TEF for 16 PAHs.

PAHs	Factors of TEF	PAHs	Factors of TEF
Nap	0.001	BaA	0.1
Acy	0.001	Chr	0.001
Ace	0.001	BbF	0.1
Flu	0.001	BaP	1
Phe	0.001	Icdp	0.1
Ant	0.01	DahA	1
Fla	0.001	BghiP	0.01
Pyr	0.001	BkF	0.1

where IR is ingestion amount of allium fistulosum; CF is the conversion factor that is used to convert fresh green vegetable weight to dry weight; BW is the average body weight [22-23].

The incremental lifetime cancer risks (ILCRs) via vegetable ingestion is calculated using Equation (3) [21]:

$$\text{ILCRs} = (\text{ED} \times \text{EF} \times \text{ED} \times \text{SF} \times \text{CF})/\text{AT} \quad (3)$$

where EF is the exposure frequency; ED is the exposure duration; SF is cancer slope factor of B[a]P; CF is a conversion factor; AT is the average life span [24].

All the reference values for Equations (2) and (3) are represented in Table 2.

Results and Discussion

Concentrations of PAHs in Allium Fistulosum

The total concentrations of 16 PAHs in home-grown allium fistulosum near deep-processing petroleum industry ranged from 87.8 to 274.78 ng/g (Table 3) with a mean concentration of 162.38 ng/g in edible parts, and from 159.01 to 430.22 ng/g with a mean concentration of 264.75 ng/g in roots. The concentrations of 16 PAHs in roots were generally higher than in the edible parts. The results suggest that adsorption of PAHs through roots from contaminated soils is an important pathway for accumulation of pollutants in vegetables [25-26].

All 16 PAH congeners were detected in allium fistulosum. As presented in Table 4, in edible parts, high-ring PAHs (>4) were predominant, accounting for 57% of the total 16 PAHs, the proportions of BaP and DahA were the highest, 22% and 16% respectively; whereas the low-ring PAHs (≤4) were accounted for 23% (<4) and 20% (4-ring) of the total 16 PAHs. In the roots, high-ring PAHs (>4) were predominant, accounting for 57% of the total 16 PAHs, the proportions of BaP and DahA were the highest, 19% and 13% respectively. Previous studies indicate that because of their greater water solubility, volatility and bioavailability, low-ring and 4-ring were predominant in 6 types of vegetables

Table 2. The reference values for equations (2) and (3).

Reference	Children		Teenagers		Adults		Old	
	Male	Female	Male	Female	Male	Female	Male	Female
IR (g/d)	95	90	160	140	240	220	230	200
EF (d/a)	365	365	365	365	365	365	365	365
ED (d)	7	7	7	7	43	43	10	10
SF (mg/kg d)	7.3	7.3	7.3	7.3	7.3	7.3	7.3	7.3
CF (mg/ng)	10 ⁻⁶							
BW (kg)	25.13	23.96	49.04	46.26	65.07	55.58	63.17	54.27
AT (d)	25500	25500	25500	25500	25500	25500	25500	25500

Table 3. Descriptive statistics of 16 PAHs concentrations in allium fistulosum.

PAHs	Edible part (ng/g)		Root (ng/g)	
	Min-Max	Mean	Min-Max	Mean
Nap	1.53-19.37	6.35	2.37-24.77	9.83
Ace	0.79-19.04	5.52	0.47-30.03	9.42
Acy	0.51-15.47	5.83	1.03-24.64	9.93
Flo	1.22-12.76	4.44	1.42-23	7.36
Phe	1.04-17.46	8.05	2.53-26.43	12.08
Ant	0.49-21.85	7.11	0.85-30.35	11.01
Fla	2.48-22.12	9.14	7.23-33.84	15.00
Pyr	1.17-14.21	6.83	2.04-20.94	12.50
BaA	1.14-16.82	7.61	2.04-20.94	11.25
Chr	0.45-22.53	8.33	2.93-32.83	14.77
BbF	0.47-18.9	8	2.65-30.3	14.61
BkF	3.17-22.23	13.17	17.89-38.75	27.84
BaP	27.3-57.17	34.93	35.15-92.1	50.53
DahA	15.6-41.19	26.25	18.7-45.38	35.71
IcdP	1.06-16.97	6.21	5.77-22.6	12.06
BghiP	1.35-9.4	4.52	4.25-25.2	10.87
∑PAHs	87.8-274.78	162.38	159.01-430.22	264.75

[6]. Different from previous studies, the high-ring PAHs in allium fistulosum grown in the study area were the predominant, this may be attributed to local anthropogenic pollution, affecting accumulation of PAH congeners in the allium fistulosum.

Spatial Distribution of PAHs in Allium Fistulosum

The spatial distribution of total 16 PAHs concentrations in edible parts of home-grown allium fistulosum are given in Fig. 2. Among the nine sampling sites, SQ, HG, JS and JH had higher PAHs

concentrations, the values were higher than 200 ng/g. HG and SQ are located around the deep-processing petroleum industry, and could be significantly affected by atmospheric PAH pollution. Deep-processing petroleum industry produces a large amount of PAHs, and may contaminate surrounding environment, influencing the accumulation of PAHs in vegetables [27]. Therefore, the concentrations of PAHs in edible parts of allium fistulosum were much higher at sampling sites SQ, HG, JS and JH. As expected, the lower concentrations of PAHs in edible parts of allium fistulosum were found at the sampling sites (SJ, TT, ZJ, WX and YL), since sampling site ZJ, WX and YL

Table 4. Proportions of individual PAHs in allium fistulosum.

PAHs	Edible parts (percentage)	Root (percentage)
Nap	3.91%	3.71%
Ace	3.40%	3.56%
Acy	3.59%	3.75%
Flo	2.73%	2.78%
Phe	4.96%	4.56%
Ant	4.38%	4.16%
Fla	5.63%	5.67%
Pyr	4.21%	4.72%
BaA	4.69%	4.25%
Chr	5.13%	5.58%
BbF	4.93%	5.52%
BkF	8.11%	10.52%
BaP	21.51%	19.08%
DahA	16.17%	13.49%
IcdP	3.82%	4.56%
BghiP	2.78%	4.11%
Low-ring	22.97%	22.52%
4-ring	19.65%	20.21%
High-ring	57.32%	57.27%

are far from the deep-processing petroleum industry, and sampling sites SJ, TT are located in the other side of Songhua River from the major PAH pollution source.

As expected, SQ, HG, JS and JH located around the deep-processing petroleum industry had higher PAH concentrations. HG and SQ located near major pollution source were much higher than at sampling sites JS

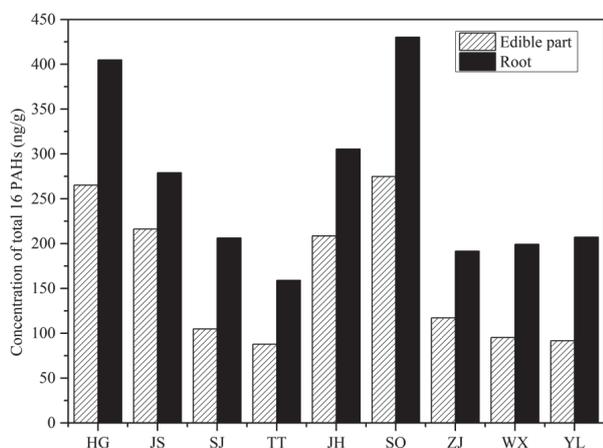


Fig. 2. The concentrations of total 16 PAHs in allium fistulosum from different sampling sites.

and JH, the concentrations were higher than 400 ng/g. The sampling sites (SJ, TT, ZJ, WX and YL) are located far from the deep-processing petroleum industry, the concentrations of PAHs in the roots of allium fistulosum were much lower.

The spatial distribution pattern of PAHs in edible parts is similar with PAHs in the roots, the high PAHs concentrations in home-grown allium fistulosum occurred in the sampling sites located around the deep-processing petroleum industry.

Source Identification of PAHs in Allium Fistulosum

The PAH isomer ratios have been employed to identify the sources of PAHs in different environmental compartments [28]. In this study, as shown in Fig. 3, the Ant/(Ant + Phe) ratios for edible parts ranged from 0.08 to 0.71, for roots ranged from 0.1 to 0.76, the most of which were higher than 0.1. The BaA/(BaA + Chr) ratios for edible parts ranged from 0.36 to 0.96, for roots ranged from 0.28 to 0.83, the most of which ranged were higher than 0.35. The results suggest that combustion of petroleum, coal and biomass were the major sources of PAHs in allium fistulosum [29-30].

The PAH isomer ratios suggest that the sampling sites are located near deep-processing petroleum industry, therefore the most of sampling sites had PAHs pollution generated from petroleum and coal combustion. In addition, the most of sampling sites had PAHs pollution derived from biomass combustion, since burning agricultural wastes took place in the study area.

Exposure and Cancer Risks of PAHs in Allium Fistulosum

This study estimated daily dietary exposures of consumers to PAHs via home-grown edible parts

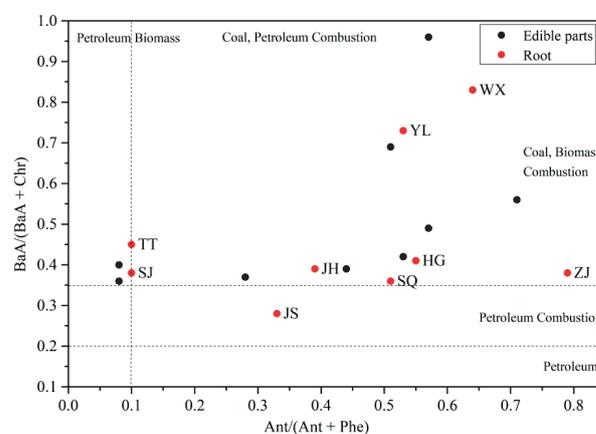


Fig. 3. PAH cross-plots for the ratios of Ant/(Ant + Phe) vs. BaA/(BaA + Chr).

Table 5. E_D (ng/(d·kg)) of PAHs in edible parts of allium fistulosum from different sampling sites.

Sampling site	Children		Teenagers		Adults		Old	
	Male	Female	Male	Female	Male	Female	Male	Female
HG	2.07E-04	2.06E-04	1.79E-04	1.66E-04	2.02E-04	2.17E-04	1.99E-04	2.02E-04
JS	2.63E-04	2.62E-04	2.27E-04	2.11E-04	2.57E-04	2.76E-04	2.54E-04	2.57E-04
SJ	9.65E-05	9.59E-05	8.33E-05	7.73E-05	9.42E-05	1.01E-04	9.30E-05	9.41E-05
TT	8.76E-05	8.71E-05	7.56E-05	7.02E-05	8.55E-05	9.18E-05	8.44E-05	8.54E-05
JH	2.31E-04	2.29E-04	1.99E-04	1.85E-04	2.25E-04	2.41E-04	2.22E-04	2.25E-04
SQ	2.54E-04	2.52E-04	2.19E-04	2.03E-04	2.48E-04	2.66E-04	2.45E-04	2.48E-04
ZJ	1.43E-04	1.42E-04	1.24E-04	1.15E-04	1.40E-04	1.50E-04	1.38E-04	1.40E-04
WX	1.32E-04	1.31E-04	1.14E-04	1.06E-04	1.29E-04	1.38E-04	1.27E-04	1.29E-04
YL	1.21E-04	1.20E-04	1.04E-04	9.65E-05	1.18E-04	1.26E-04	1.16E-04	1.17E-04
Average	1.71E-04	1.69E-04	1.47E-04	1.37E-04	1.66E-04	1.79E-04	1.64E-04	1.66E-04

Table 6. ILCRs of PAHs in edible parts of allium fistulosum from different sampling sites.

Sampling site	Children		Teenagers		Adults		Old	
	Male	Female	Male	Female	Male	Female	Male	Female
HG	4.2E-06	4.2E-06	3.7E-06	3.4E-06	2.5E-05	2.7E-05	5.8E-06	5.9E-06
JS	5.4E-06	5.4E-06	4.7E-06	4.3E-06	3.2E-05	3.5E-05	7.4E-06	7.5E-06
SJ	2.0E-06	2.0E-06	1.7E-06	1.6E-06	1.2E-05	1.3E-05	2.7E-06	2.8E-06
TT	1.8E-06	1.8E-06	1.5E-06	1.4E-06	1.1E-05	1.2E-05	2.5E-06	2.5E-06
JH	4.7E-06	4.7E-06	4.1E-06	3.8E-06	2.8E-05	3.0E-05	6.5E-06	6.6E-06
SQ	5.2E-06	5.2E-06	4.5E-06	4.2E-06	3.1E-05	3.3E-05	7.2E-06	7.2E-06
ZJ	2.9E-06	2.9E-06	2.5E-06	2.3E-06	1.8E-05	1.9E-05	4.0E-06	4.1E-06
WX	2.7E-06	2.7E-06	2.3E-06	2.2E-06	1.6E-05	1.7E-05	3.7E-06	3.8E-06
YL	2.5E-06	2.5E-06	2.1E-06	2.0E-06	1.5E-05	1.6E-05	3.4E-06	3.4E-06
Average	3.5E-06	3.5E-06	3.0E-06	2.8E-06	2.1E-05	2.2E-05	4.8E-06	4.9E-06

of allium fistulosum consumption are shown in Table 5. In terms of gender and age groups, the daily exposure doses of PAHs via allium fistulosum consumption ranged from 2.76×10^{-4} to 7.02×10^{-5} ng/(d·kg). Among four age groups, children had the highest daily exposure dose, while teenagers had the lowest. This results are attributed to the differences in intake amounts and body weights of four age and gender groups [6].

The ILCRs was applied to assess the potential cancer risks to consumers exposed to PAHs via the consumption of edible parts of home-grown allium fistulosum. The values of ILCRs lower than 10^{-6} are indicated a safe risk level, the values ranged 10^{-6} - 10^{-4} are considered a low risk, while the values of ILCR greater than 10^{-4} predict a high potential risk [31-32]. As shown in Table 6, the ILCRs values of edible parts of allium fistulosum for different gender and age groups

were ranged from $1.4E-06$ to $3.5E-05$, which suggest low risks.

According to the results for ILCRs, in four age groups, the adults had the highest cancer risk exposed to PAHs via home-grown allium fistulosum consumption, while the teenagers had the lowest cancer risk. The results suggest that local adults are more susceptible to PAHs in edible parts of home-grown allium fistulosum. For different sampling sites, cancer risks exposed to the edible parts of allium fistulosum sampled from JS and SQ were the highest for children, teenagers, adults and old people, while cancer risks exposed to the allium fistulosum sampled from TT and SQ were the lowest. The risk levels were not consistent with the concentration levels of 16 PAHs, this may be attributed to the differences in toxic effects and concentrations of 16 individual PAHs.

Conclusions

The concentrations of total 16 PAHs in edible parts of *Allium fistulosum* ranged from 87.8 to 274.78 ng/g with a mean concentration of 162.38 ng/g, and total 16 PAHs in roots ranged from 159.01 to 430.22 ng/g with a mean concentration of 264.75 ng/g. For individual PAHs, the concentrations of high-ring PAHs were the highest. Adsorption via the roots and local anthropogenic pollution were found to be determinants of accumulation of PAHs in edible parts of *Allium fistulosum*. The sources of PAHs were identified using PAH isomer ratios, the results suggest that the main sources of PAHs were vehicle emissions, agricultural waste burning and industrial emissions. The study of daily dietary exposures indicates that consumers should reduce the consumption of home-grown *Allium fistulosum* from the urban areas of industrial city. The results for ILCRs suggest that the PAHs in home-grown *Allium fistulosum* might pose a low cancer risk, and female adults had the highest cancer risk. In addition, home-grown *Allium fistulosum* from JS and SQ posed higher cancer risks to children, teenagers, adults and old.

Acknowledgments

This work was supported by the Major Project of Science and Technology for Planning and Development (No. 20200403019SF), and Major project of Science and Technology of Jilin Institute of Chemical Technology (No. 2018025).

Conflict of Interest

The authors declare no conflict of interest.

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