SOURCE APPORTIONMENT ANALYSES OF CADMIUM AND ARSENIC IN TYPICAL PERI-URBAN VEGETABLE SOIL OF XIJIANG RIVER BASIN: A CASE STUDY OF ONE GROWTH CYCLE OF VEGETABLES ON A SMALL SCALE

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Abstract. We explored pollution input-output of soil Cd and As for one growth cycle of vegetables in peri-urban vegetable soil along Xijiang river, preliminarily verified the input-output of IW, discussed source apportionment of Cd and As in PFs, RW and IW, systematically calculated Cd and As contents in local PFs, IW and RW. The conclusions were as follows, IW had the largest effect on Cd input to the vegetable soil, mean content was 0.1050 $mg\cdot(m^2)^{-1}$ in the growth cycle, followed by PFs and RW. PFs had the largest effect on As input to vegetable soil, mean content was 2.300 $mg\cdot(m^2)^{-1}$, followed by RW and IW. According to the input-output of IW and preliminary incomplete statistical analyses, Cd in IW fully showed input status and As fully showed output status to the vegetable soil, suggested to increase the risk monitoring of Cd in IW and As in PFs.
Keywords: *Cd, As, irrigation water (IW), pesticides and fertilizers (PFs), rainwater (RW)*

Introduction

With the rapid development of the local economy, some farmlands have gradually evolved into peri-urban farmlands and the pollution risks of the external environment have increased significantly. Soil contamination by heavy metals is a serious global environmental problem (Husejnovic et al., 2018), compound pollution by cadmium (Cd) and arsenic (As) is a typical form of farmland contamination by heavy metals, which mainly originates from pesticides and fertilizers (PFs), irrigation water (IW) and rainwater (RW) in the Pearl River Delta (Zhou et al., 2014; Zhang et al., 2019). And the effects of phosphate fertilizer and nitrogen fertilizer are to improve yellow leaves, fruit expansion, coloring, increasing yield, flowering, rooting, strengthening stems and so on. The nutrients required for vegetables, are mainly from base fertilizer, topdressing fertilizer and foliar fertilizer, sufficient soil nutrients are necessary for the growth of vegetables, and they also have potential high risk of Cd and As. Source apportionment research of Cd in the paddy soil found that IW was the largest input item, which was higher than the annual input of phosphate fertilizer, organic fertilizer and atmospheric sedimentation (He et al., 2016). Some scholars (Arshadi et al., 2011; Xu et al., 2017a) have pointed out that sharply reducing Cd in IW was an effective way to achieve the

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reduction of Cd levels in the vegetable soil and safe production of rice (*Oryza sativa* L.). Natural and anthropogenic sources have led to high As in rice grains, it was also very important to strengthen the short-term studies of the paddy soil pollution by As in the water systems (Kumarathilaka et al., 2018). A study using features of the five rainfall events in July and August of 2019 revealed the temporal and spatial variations of surface pollution (Yang et al., 2021), it can be seen that data analyses of short-term research were also very important.

In recent years, many scholars have carried out source apportionment analyses of soil pollution by heavy metals, while the research in typical peri-urban vegetable soil was lacking. It was foreseeable that the continuous cultivation of the vegetable soil may have higher ecological risks of Cd and As under the high-frequency irrigation and IW flooding. We visited the local farmers and investigated the actual situation in the production, made a quantitative estimation of Cd and As sources of the vegetable soil for one growth cycle. The annual base fertilizer was applied in July and August, so the study of the growth cycle can reflect maximum input of Cd and As into soil. In other words, the growth cycle was the biggest pollution cycle and it also fed back the biggest pollution cycle of anthropogenic source in the next whole year. And the studied area was the best representative and was the same as the production mode of many farmlands along the river in the lower reaches of Xijiang River. The results provided supplementary data for prevention and remediation of soil contamination by Cd and As in the typical farmland (Ren et al., 2022).

Materials and methods

Studied area

Studied area is located in Xijiang Park, Foshan City, China. The vegetable soil along the river was in a state of normal cultivation. *Figure 1* showed water sampling points, included FSJS1 to FSJS7, and the sampling points were water intake points for direct irrigation of the vegetable soil along the river. There were two typical irrigation modes by the groundwater and the river. There were many kinds of PFs, and local vegetables were mainly planted with leafy vegetables (*flowering Chinese cabbage*, *Indian lettuce*, *romaine lettuce*, *cabbage*, etc.) and fruit vegetables (*white gourd*, *tomato*, *cowpea*, *sweet corn*, *okra*, etc.), one growth cycle of leafy vegetables ranged from about 2 to 3 months and fruit vegetables ranged from about 2 to 6 months. We collected related samples from late July to mid-November 2019, the research period covering one growth cycle was important, the growth cycle in the study was 4 months, July was at the end of farming slack rather than the growth cycle, August was in the seedling stage, September was in the growth stage and November was in the maturity stage. Some metals in vegetable soil in July were removed by vegetable soil drainage from RW runoff, base fertilizer in the soil was almost completely consumed. And new base fertilizer would be applied again at seedling stage.

We have studied distribution characteristics of total Cd and total As (*Figure 2*) in river sediment, riverside farmland soil and inside-dyke farmland soil. *Figure 2* showed content distribution of total Cd and total As in river sediment and farmland soil (Ren et al., 2022). As showed in *Figure 1*, river sediment was marked as FSJS1 to FSJS7, riverside farmland soil was marked as FSJ2Fa to FSJ7Fb, inside-dyke farmland soil was marked as FSX1Fa to FSX3F. Research results at the end of the farming slack reflected minimum contents of soil contamination by Cd and As in the farmland, which affected

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by anthropogenic sources since the application of topdressing last autumn, and can fully mirror the distribution and development of Cd and As in the farmland (Ren et al., 2022).

Figure 1. *Sampling site layout and surrounding environment in the studied area*

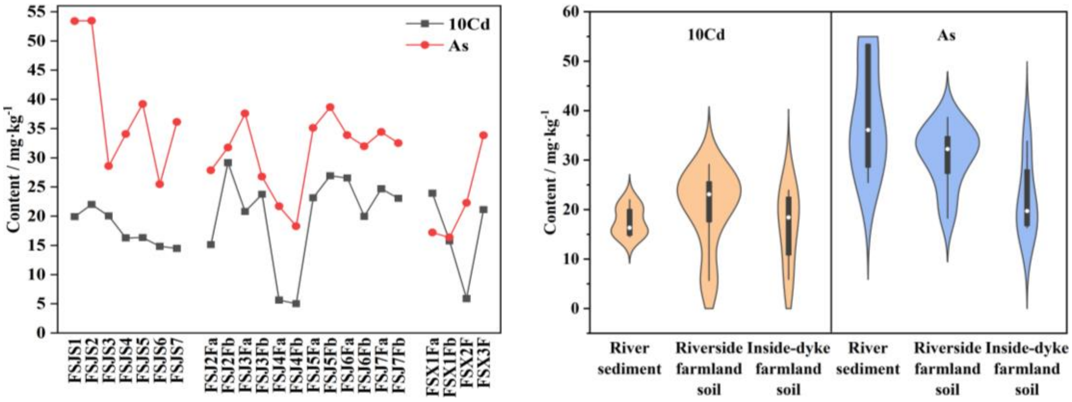

Figure 2. *Content distribution of Cd and As in the river sediment and the farmland soil*

Models and formulas

Based on export coefficient models, we used the unit load approach, which measured produced content of pollutants in each calculation unit (Liu et al., 2015). Rain is the most important part of local atmospheric deposition in the rainy season, its migration in the surface environment is mainly RW. Local annual rainfall days ranged from 110 to 190 days, the studied area was about 150 days, and about 80% was moderate rain and heavy rain (Zheng et al., 2017). Extreme weather was frequent during the investigation; the traditional long-term sampling method was difficult.

Figure 3 showed the layout of RW sampling points, the samples were collected in a roof, having five catchment areas, each catchment area was about 0.35 m^2, getting a mixed 1.2 L sample by five catchment areas. There were five RW samples named RW1-RW5. The content of pollutants decreased gradually with the increase of rainfall time, pollutants were concentrated at the beginning of rainfall (Xu et al., 2017b), the samples were collected at the beginning of rainfall in the investigation.

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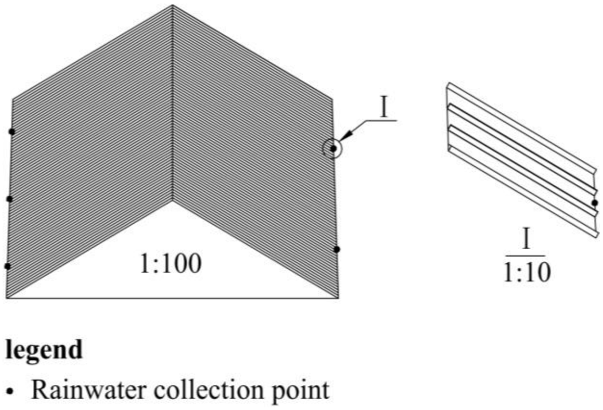

Figure 3. *Layout of RW sampling points*

The rainfall for July averaged 223.7 mm, August averaged 319.1 mm, September averaged 100.6 mm, October averaged 44.0 mm and November averaged 0.3 mm in Guangdong province (Foshan, 2019), total rainfall for one growth cycle was 464.0 $L \cdot (m^2)^{-1}$. The calculation formula of input content of metal from RW is as follows:

$$y \approx a \cdot b \cdot 10^{-3} \quad \text{(Eq.1)}$$

where y is input content of metals from RW of one growth cycle, $mg \cdot (m^2)^{-1}$; a is the mean content of metals in RW, $\mu g \cdot L^{-1}$; b is the total rainfall for one growth cycle, $L \cdot (m^2)^{-1}$.

The local rainfall is abundant, the temperature is very high and irrigation is also frequent from July to November. More RW will increase Cd and As to move into local river, meanwhile the part of Cd and As can come back the vegetable soil by the frequent irrigation, which is input-output of Cd and As in IW supply and drainage. IW volume was calculated referring to "*Norm of water intake Part 1: Agriculture (DB44/T 1461.1-2021)*", studied area belonged to sub-area of IW norm on the Pearl River delta plain in central Guangdong. The guarantee rate of hydrological year selected 75%, norm water intake of IW for leaf vegetables was 0.21 $m^3 \cdot (m^2)^{-1}$, norm water intake of IW for fruit vegetables was 0.54 $m^3 \cdot (m^2)^{-1}$. The calculation formulas of the dynamic content of one growth cycle, net input contents of IW are as follows:

$$y \approx A \cdot \Delta \cdot 10^{-3} \quad \text{(Eq.2)}$$

$$\Delta \approx R - B \quad \text{(Eq.3)}$$

$$I = 1000 \times c \cdot d \quad \text{(Eq.4)}$$

where y is the dynamic content of metals for one growth cycle in water samples, $mg \cdot (m^2)^{-1}$; A is IW volume for a specific soil area, $L \cdot (m^2)^{-1}$; Δ is net input content of metals in IW, $\mu g \cdot L^{-1}$. R means the average content of metals in the farming slack period, $\mu g \cdot L^{-1}$. B means the mean content of metals in the farming busy period, $\mu g \cdot L^{-1}$. I is IW volume, L. c is norm water intake of IW, $m^3 \cdot (m^2)^{-1}$. d is area, m^2.

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Sampling

Firstly, we selected the appropriate sampling time through the weather forecast published by China Weather Network. Secondly, we used mobile tool to assist sampling, completed the sampling point layout through Aowei Map (a Chinese mobile positioning APP) and took sampling according to the positioning information. Thirdly, water samples needed to be stored at low temperature during transportation, were mixed and divided into two parts in the laboratory, stored in a refrigerator at 4 °C, completed the pretreatment as soon as possible and the sample detection within the specified time. Finally, each sample tested three times, average value (± standard deviation) was used for analyses of heavy metals in all samples.

Reagents and instruments

Total Cd in water samples takes the digestion method of HNO_3-$HClO_4$ of mixed acid, using atomic absorption spectrometer (Pin AAcle 900T, PerkinElmer, USA). Total As in water samples takes the digestion method of sample pretreatment, referring to Chinese standard of *"Monitoring and analysis methods for water and wastewater"* (China, 2002), using atomic fluorescence spectrometer (AFS-9700, Beijing Haiguang Instrument Co., Ltd., China). Dissolved part contents of Cd and As refer to the part that can pass through the 0.45 μm filter membrane in the raw water, referring to Chinese standard (China, 2002).

The experimental reagents are all high grade pure. The laboratory utensils are soaked in 10% nitric acid for more than 24 h, and are washed with high-purity water before use. Reagent blank and 20% parallel samples are made for each batch of samples in the experiment. The recovery rate of heavy metal blank or sample standard addition is controlled within 10%. R^2 of the standard curve of detection items is higher than 0.995, and the data accuracy meets the requirements.

Data processing

The statistical analyses of experimental data and the production of data graphs are made based on Microsoft Excel 2016, SPSS 22.0 and Origin 9.0.

Results and Discussions

PFs input study

According to the using condition of the PFs in the vegetable soil, there were four brands of compound (organic) fertilizer, two brands of foliar fertilizer and several pesticides including *pyridaben*, *avermectin*, *chlorantraniliprole*, *glufosinate*, *glyphosate* and so on, most pesticides were in bags varied from 5 to 20 g, took 1 g for each sample, mixed, grounded, sieved and kept sealed. Several straw ashes (grass ashes) were collected from the vegetable soil, while organic compost was not common in the studied area, including excrement, animal and plant residues and metabolites. *Table 1* showed Cd and As contents of local PFs, according to Chinese standard limits of "*Ecological Indicators of Arsenic, Cadmium, Lead, Chromium and Mercury in Fertilizers (GB/T 23349-2009)"*, Cd upper limit was 10 $mg\cdot kg^{-1}$ and As was 50 $mg\cdot kg^{-1}$, it can be seen that Cd and As did not exceed the standard.

Table 1 showed Cd and As contents of local PFs. Total input was estimated by the minimum value, base fertilizer and topdressing used SDL, the straw ash was calculated

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at 0 g·(m^2)$^{-1}$, the pesticide was calculated at 25 mg·(m^2)$^{-1}$ for one growth cycle. Total input was estimated at the maximum value, base fertilizer used NSDK, topdressing used JZ and SH, the straw ash was calculated at 25 g·(m^2)$^{-1}$, the pesticide was calculated at 25 mg·(m^2)$^{-1}$. Total Cd input of local common PFs ranged from 0.0005 (0.002×0.06 + 0.002×0.04×4 + 0.003×0.0001×2 + 0.035×0.0001×12 + 0.490×0.000 + 0.049×0.000025) to 0.1827 (0.154×0.07 + 0.335×0.07×2 + 0.061×0.01×4 + 0.003×0.0001×2 + 0.035×0.0001×12 + 0.490×0.250 + 0.049×0.000025) mg·(m^2)$^{-1}$, total As input ranged from 0.404 (1.835×0.06 + 1.835×0.04×4 + 0.761×0.0001×2 + 0.336×0.0001×12 + 6.995×0.000 + 3.875×0.000025) to 4.196 (8.024×0.07 + 13.358×0.07×2 + 0.374×0.01×4 + 0.761×0.0001×2 + 0.336×0.0001×12 + 6.995×0.250 + 3.875×0.000025) mg·(m^2)$^{-1}$.

Table 1. *Cd and As contents of local PFs*

Item	Brand	Cd /mg·kg^{-1}	As /mg·kg^{-1}	Total nutrients % ≥	Base fertilizer /kg·(m^2)$^{-1}$	Topdressing /kg·(m^2)$^{-1}$	Frequency of topdressing
Compound (organic) fertilizers	SDL	0.002±0.000	1.835±0.564	54	0.06-0.09	0.04-0.06	month
	YR	0.001±0.000	3.196±0.435	45	0.07-0.10	0.03-0.07	month
	BL	0.043±0.004	2.736±0.319	45	0.09-0.12	0.03-0.07	month
	NSDK	0.154±0.027	8.024±0.961	45	0.04-0.07	0.03-0.04	month
Phosphate fertilizers	JH	0.384±0.024	5.482±0.808	12	0.04-0.07	0.01-0.03	month
	JZ	0.335±0.024	13.358±1.404	12	0.07-0.15	0.04-0.07	seedling and growth
Urea	SH	0.061±0.022	0.374±0.106	46	0.01	0.01	month
	FX	0.088±0.006	0.147±0.064	46	0.01	0.01	month
Foliar fertilizers	CN	0.003±0.001	0.761±0.143	98	/	0.0001	seedling and growth
	XHEK	0.035±0.004	0.336±0.115	46	/	0.0001	10 days
Straw ashes	/	0.490±0.101	6.995±1.022	/	/	/	/
Pesticides	/	0.049±0.007	3.875±1.267	/	/	/	/

Note: the brand is an acronym

RW input study

Table 2 showed input content of Cd and As from RW. Combined with the correlation analyses between the content of Cd and As and rainfall cycle, it can be seen that there was no obvious linear relationship, the metal content in RW had little correlation with the sampling period, which may be related to the short dry period (Zhang et al., 2014). Local rainfall was heavy, and the duration of the individual rainfall was relatively long, wet deposition of heavy metals was concentrated in the pre-mid periods or even faster. According to *Eq.1*, its value was the mean content of Cd and As. Using 30% of mean content to calculate the minimum input, input content of Cd from RW ranged from 0.0033 to 0.0111 mg·(m^2)$^{-1}$, input content of As from RW ranged from 0.7534 to 2.511 mg·(m^2)$^{-1}$.

IW input study

In this study, IW referred to river water, IW was one of the main sources of Cd and As input to the vegetable soil, and it was also the main carrier for outward migration of Cd and As in the vegetable soil. Drainage was an effective mean of self-purification for the vegetable soil. IW in the studied area was greatly affected by agricultural activities

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due to terrain and other factors, and PFs mainly affected IW in the form of drainage. *Figure 3* showed Cd and As distribution in IW in July, August, September, and November. July was the end of farming slack, IW after rainfall can be regarded as the most unfavorable value of water quality under natural disturbance, Cd was 0.28 $\mu g \cdot L^{-1}$, As was 3.80 $\mu g \cdot L^{-1}$. According to *Eq.3*, $\triangle$Cd was 0.25 $\mu g \cdot L^{-1}$, $\triangle$As was -24.28 $\mu g \cdot L^{-1}$.

Table 2. *Input content of Cd and As from RW*

Sample No.	Cd / $\mu g \cdot L^{-1}$	As / $\mu g \cdot L^{-1}$	Rainfall cycle / d	Cd / $mg \cdot (m^2)^{-1}$	As / $mg \cdot (m^2)^{-1}$
RW1	0.02	6.43	1	0.0093	2.984
RW2	0.03	4.67	5	0.0139	2.167
RW3	0.02	6.88	3	0.0093	3.192
RW4	nd	5.42	7	0.0000	2.515
RW5	0.05	3.66	3	0.0232	1.698
Mean	0.02±0.02	5.41±1.30	3.8	0.0111±0.0084	2.511±0.605
Minimum	/	/		0.0033±0.0025	0.753±0.182

Note: nd means less than the detection limit, and it is calculated as 0 $\mu g \cdot L^{-1}$

There was a cycle of repeated input and output of Cd and As between IW and soil. The input risk of IW was affected by irrigation drainage. The content of Cd and As was the least affected by anthropogenic sources in July, the values used to judge the input possibility of IW. According to *Eq.4*, Cd input from IW in one growth cycle ranged from 0.0588 to 0.1512 $mg \cdot (m^2)^{-1}$, As input ranged from 0.798 to 2.052 $mg \cdot (m^2)^{-1}$.

According to *Figure 4(a)*, Cd in most IW after rain was dominated by suspended particulate phase, and was dominated by soluble Cd in August, September and November. According to *Figure 4(b)*, As in most IW after the rain was dominated by suspended particulate phase, and was mainly soluble As in August, September and November. Suspended particulate phase in natural waters controlled the interaction, transportation, and biological effects of heavy metals in water bodies, a large number of Cd carried by suspended particulate phase and it was an important source of Cd pollution in IW (Li et al., 2017), which was not inconsistent with our research conclusion. When Cd content was low, proportion of dissolved part was slightly higher in IW source. One study showed that association of As contents in IW, paddy field soil and *Oryza sativa* L. with the cropping seasons, found that As content using ground water to irrigate decreased significantly from summer to winter (Biswas et al., 2016), As trend in IW was similar to that in our study.

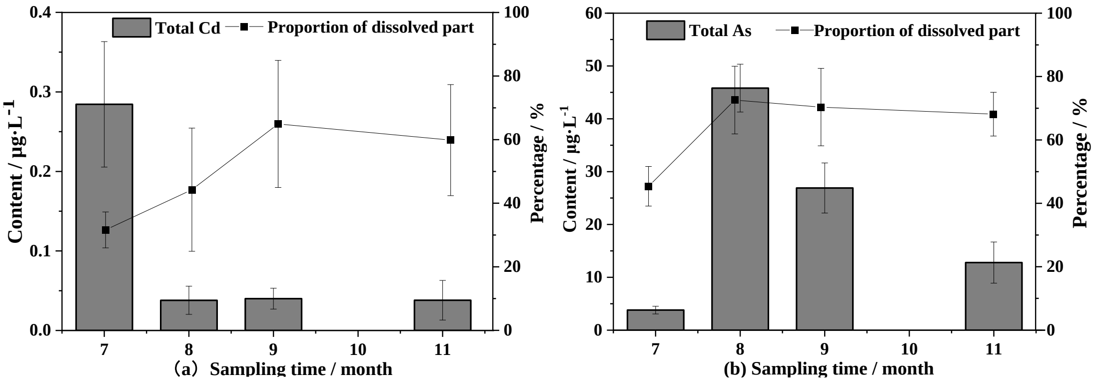

Figure 4. *Cd and As distribution of IW in July, August, September and November*

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Compared with Cd, As distribution in IW was obviously affected more by the farming cycle, irrigation and rainfall, which related to the frequency of leaching and leaching of the soil. Frequency of leaching presented roughly that August was most, followed by September, November and July. Rainfall in the Pearl River Delta region was concentrated from February to September, and water requirement for vegetable growth presented that seedling period was most, followed by growing period and mature period. Fertilizer residue in the process of vegetable growth presented that August was most, followed by September, November and July. Quantitative research preliminarily proved the input-output of Cd and As from IW.

Above, it can be judged that irrigation was an important way that Cd and As in IW from the vegetable soil after environmental self-purification (RW runoff) and irrigation drainage (*Figure 5*, Ren et al., 2021) returned to the vegetable soil by irrigation. This also explained the higher soluble Cd and As content in IW at the seedling stage and other stages.

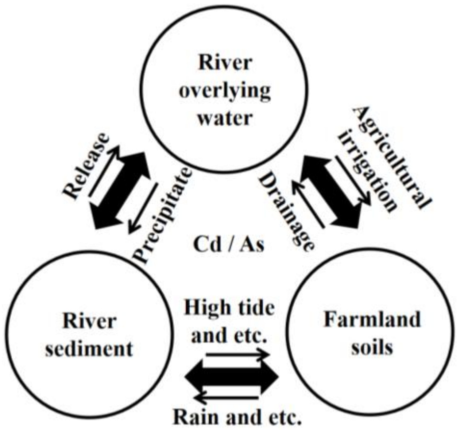

Figure 5. *Potential migration routes of Cd and As in the vegetable soil along the River*

According to *Eq.2*, dynamic input of Cd and As from IW was 0.0525 and -5.099 mg·$(m^2)^{-1}$ for leafy vegetable soil in the growth cycle. Dynamic input of Cd and As from IW was 0.1350 and -13.111 mg·$(m^2)^{-1}$ for fruit vegetable soil in the growth cycle. It can be seen that As in the vegetable soil presented output status. When the local area was frequent agricultural activities, As concentration in the IW was about 10 times higher than usual concentration, and the dynamic input content was negative, the value can reflect the dynamic process of the irrigation, the result roughly reflected the As content of agricultural sources released by local agricultural activities to the water environment.

Input of Cd and As in the growth cycle

Table 3 showed calculation results of input contents of Cd and As from PFs, RW and IW in the vegetable soil. IW had the largest impact on Cd input to the vegetable soil, followed by PFs and RW. Combining characteristics of the vegetable soil and changes in IW quality, it was found that the net input of Cd from IW showed input status. PFs had the greatest impact on As external input to the vegetable soil, followed by RW and IW, according to dynamic input potential of IW, found that As from IW showed output status. For relatively high As contents in IW, the addition of compost to the soil could

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be used as an effective means to limit vegetable As accumulation from As-contaminated water irrigation (Caporale et al., 2016). According to IW net input, it was a preliminary judgment that IW might be influenced more sources of heavy metal pollution and irrigation drainage was the most obvious reason, it might also be related to other input sources of the vegetable soil contamination by Cd and As. Besides these significant pollution sources, random input of the original soil and point sources may also be important factors affecting the results.

Table 3. *Calculation results of input contents of Cd and As*

Input item	Cd / mg·(m^2)$^{-1}$			As / mg·(m^2)$^{-1}$		
	Max.	**Min.**	**Mean**	**Max.**	**Min.**	**Mean**
PFs	0.1827	0.0005	0.0916	4.196	0.404	2.300
RW	0.0111	0.0033	0.0072	2.511	0.753	1.632
IW	0.1512	0.0588	0.1050	2.052	0.798	1.425
Total input content	0.3450	0.0626	0.2038	8.759	1.955	5.357
Net input of IW	0.1350	0.0525	0.0938	-13.111	-5.099	-9.105

Conclusions

1) Different geographical location was bound to be accompanied by different external influences. And peri-urban vegetable soil was bound to increase potential pollution risks, mainly depending on the pollution degree of IW and PFs. IW had the largest impact on Cd input to the vegetable soil, followed by PFs and RW. PFs had the greatest impact on As input, followed by RW and IW.

2) Based on the dynamic input of IW, Cd fully showed the trend of input to the vegetable soil and As fully showed the trend of output from the vegetable soil. The input risk of IW was affected by irrigation drainage, there was a cycle of repeated input and purification (output) between IW and soil, there was a continuous cycle process that the part of Cd and As can migrate into IW by RW runoff and drainage, which repeatedly returned to the vegetable soil by irrigation. This also explained the higher content of soluble Cd and As in IW at the seedling stage and other stages.

3) Local soil pollution by Cd and As had the multiple effects by the environment and agricultural activities, suggested to increase the risk monitoring of Cd in IW and As in PFs. Besides, the area should optimize treatment methods of irrigation water supply and drainage.

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Conflict of Interests. The authors declare no conflict of interests.

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