

## Exposure to PAHs and Health Risk Assessment in Three Sewage Irrigations (Pendedahan kepada PAH dan Kesihatan Penilaian Risiko dalam Tiga Pengairan Kumbahan)

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### ABSTRACT

*The sewage irrigation area in Tianjin reached 6 billion m<sup>2</sup> in 2003 which accounts for 22% of the irrigated areas. Polycyclic aromatic hydrocarbons (PAHs) contamination caused by sewage irrigation on agricultural land has attracted considerable attention because of their toxic and hazardous risks to human beings. In order to evaluate the health risk of PAHs to farmers in four age groups, a multimedia/multipathway exposure model was applied. The results showed that the chronic daily intake (CDI) of children, adolescents, adults and the aged to the 16 PAH compounds exposure were 2.83, 2.34, 1.44, 1.05  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , respectively. The non-cancer risk of life long exposure in sewage irrigation was  $2.75\times 10^{-3}$  and the cancer risk was  $2.49\times 10^{-5}$ . From the long-term safety point of view, the health risks caused by PAHs were at a relatively high level. Hence, we suggest that crops of sewage irrigated areas and their distribution channels should be re-concerned by the government in the future city planning.*

*Keywords: Farmers; health risks; sewage irrigation; PAHs*

### ABSTRAK

*Kawasan pengairan kumbahan di Tianjin mencapai 6 bilion m<sup>2</sup> pada tahun 2003, mencakupi 22% daripada kawasan pengairan. Pencemaran hidrokarbon aromatik polisiklik (PAH) yang disebabkan oleh pengairan kumbahan di atas tanah pertanian telah menarik perhatian kerana risiko toksik dan berbahaya kepada manusia. Dalam usaha untuk menilai risiko kesihatan PAH kepada petani dalam empat kumpulan umur, satu model pendedahan pelbagai laluan multimedia telah digunakan. Hasil kajian menunjukkan bahawa pengambilan yang kronik setiap hari (CDI) kanak-kanak, remaja, dewasa dan orang tua kepada pendedahan 16 sebatian PAH masing-masing adalah 2.83, 2.34, 1.44, 1.05  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ . Risiko kanser bukan-pendedahan sepanjang hayat dalam pengairan kumbahan adalah  $2.75 \times 10^{-3}$  dan risiko kanser adalah  $2.49 \times 10^{-5}$ . Daripada sudut keselamatan jangka panjang pandangan, risiko kesihatan yang disebabkan oleh PAH berada pada tahap yang agak tinggi. Oleh itu, kami mencadangkan agar tanaman kawasan pengairan kumbahan dan saluran pengedaran perlu diambil kira oleh kerajaan dalam perancangan bandar masa depan.*

*Kata kunci: PAH; pengairan kumbahan; petani; risiko kesihatan*

### INTRODUCTION

PAHs contamination caused by sewage irrigation on agricultural land has attracted considerable attention because of their carcinogenic risks to human beings. Because of shortage of water resources annually, the cropland in Tianjin has been irrigated with sewage since 1958 (Ashraf et al. 2013; Yang et al. 2005). It was shown that sewage irrigation area of Tianjin reached 6 billion m<sup>2</sup> in 2003, which accounted for 22% of the total irrigated area of the city (Ashraf et al. 2014; Qi & Wu 2012; Zhong & Li 2011). There are three sewage irrigations areas in Tianjin: South River sewage Irrigation, Beitang River sewage Irrigation and Beijing River sewage Irrigation.

Polycyclic aromatic hydrocarbons (PAHs) are one of the most important contaminations in sewage, which are listed as priority environment pollutants in China and U.S EPA (Ashraf et al. 2012a; Lu et al. 2011). PAHs are compounds which contain two or more than two benzene rings. They are also persistent organic pollutants with high refractory, toxicity and carcinogenicity (Alficia et al. 2012;

Ashraf et al. 2012b; Luo et al. 2005). PAHs mainly come from oil refining, incomplete combustion of fossil and coal. PAHs migrate through the dry/wet deposition, sludge and sewage irrigation channels into the soil, where the pollution is gradually accumulated (Ashraf et al. 2012c; Wilcke 2000). PAHs can enter the plants by the root absorption from soils or the atmosphere sedimentation and can accumulate and migrate in plants, which does harm to human health through the food chain (Ashraf et al. 2012d; Wan et al. 2009). Exposure assessment is an important health risk assessment process. Hence, this study selected multimedia/multipathway exposure model as the main method for health risk assessment. By applying the exposing model, we can quantitatively estimate the absorption of PAHs to the human body through various pathways (Ashraf et al. 2012e; Muhammad & Salam 2012; Nam et al. 2008).

Farmers are in direct contact with pollution sources, which makes them one of the direct indicators of the health risks of harmful pollutants (Ashraf et al. 2011; Ni et al. 2012). Up to date, there is very limited knowledge

available on the impact of PAHs pollution on the health of agricultural workers. Therefore, the current study selected sewage irrigated agricultural workers as the study group, and estimated the exposure and the health risks of PAHs of agricultural workers through exposure model. The results would be important for quantitative research on the impact of pollutants on human health. It would also provide important scientific guidelines for the decision making of environmental managers.

#### METHODS AND MATERIALS ORGANIC MATTERS ANALYSIS

The samples were collected from 20 sewage irrigation sites in Tianjin (Figure 1). Water, soil and crop samples were taken at each sampling site. In total 160 samples were collected, including 46 water samples, 92 soil samples and 37 crops samples (including 17 grains and 20 vegetables).

PAHs in water samples were analyzed using the method based on GB HJ 478-2009. Firstly, 1 L sample was extracted, purified and steamed. Secondly, nitrogen blowing was used to reduce the volume in 1 mL. The samples were then stored at 4°C in 2 mL brown bottle and seal stamped with Para film until analysis. PAHs concentrations were tested with HPLC using gradient elution: acetonitrile 65% and water 35%, 27 min; increments on 2.5% acetonitrile/min to 100% acetonitrile until the peak was completed. PAHs in soil samples were analyzed using the method documented in EPA 3540(3). The soil samples were freeze-dried, grounded with a mortar into powders and filtered through 100-mesh sieve. The samples were then analyzed using the same procedures as the water samples.

Crop samples were pre-treated using the ultrasonic extraction method (Ashraf et al. 2010; Qin et al. 2010). Firstly, the crop samples were weighted, labeled and

transferred to a beaker. Secondly, dichloromethane was added and the samples were ultrasonic twice for 60 min at 40% power. The extracts were combined after filtration funnel, separating the organic phase extract. The samples were then treated the same as the water samples.

#### MATERIALS AND METHODS

Due to differences in behavioral parameters such as the breathing speed and food consumption in different age groups, the population is divided into four groups according to age (i.e. 0-6 years old children (sensitive populations); 7 to 18 year old adolescent (moderately sensitive populations); adults aged 19 to 55 and 55 years or older. Compared with other residents in Tianjin, irrigation workers were exposed to more pollutants. Hence, we calculated seven possible routes of exposure, respectively (including ingestion of dust, grains and vegetables, breathing, drinking water, dust exposure and skin dermal exposure). The actual exposure, specific subsets of the total daily exposure, specific subsets of unit weight daily exposure and lifetime daily exposure were calculated as well.

The multimedia/multipathway exposure model of U.S EPA was applied (U.S. EPA 1997). The chronic day ingestion (CDI) dose was calculated using the following calculation formula (Feng et al. 2011; Li et al. 2009, 2005):

$$CDI_{ij} = \frac{CS_i \times IR_{ij} \times CR_{ij}}{BW_i} \times \frac{EF \times ED}{AT} \times CF \quad i = 1 \dots 3, j = 1 \dots 7, \quad (1)$$

where  $CDI_{ij}$  is the unit weight daily exposure of  $i$  subsets by  $j$ -exposure pathways;  $\text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ ;  $CS_i$  is the contaminant levels of  $i$  medium;  $\text{mg} \cdot \text{kg}^{-1}$  or  $\text{mg} \cdot \text{L}^{-1}$ ;  $IR_{ij}$  is the uptake rate;



FIGURE 1. Sampling points in Tianjin Sewage Irrigation

$\text{mg}\cdot\text{d}^{-1}$  or  $\text{ml}\cdot\text{d}^{-1}$ ;  $CR_{ij}$  is the absorption factor, dimensionless;  $CF$  is the conversion factor,  $10^{-6}$ ,  $\text{kg}\cdot\text{mg}^{-1}$ ;  $BW_i$  is the body weight, kg;  $ED$  is the exposure year, a;  $EF$  is the exposure frequency,  $\text{d}\cdot\text{a}^{-1}$ ; and  $AT$  is the total exposure time, day.

The study calculated the value of all the required parameters reported in the literature (Feng et al. 2011). In adults, for example,  $EF$  is  $180 \text{ d}\cdot\text{a}^{-1}$ ,  $ED$  is 30a (non-carcinogens) or 70a (carcinogens);  $BW$  is 70 kg;  $AT$  is 10950d (non-carcinogens) or 25550d (carcinogens);  $CR$  is  $16600\text{cm}^2$  or  $5000 \text{ cm}^2$  (soil contact); Skin absorption parameters is  $0.001 \text{ cm}\cdot\text{h}^{-1}$ ; average life expectancy is 75 years.

Risk into cancer includes carcinogenic risk and non-carcinogenic risk (Bakar et al. 2013; Maria et al. 2013; U.S. EPA 1989). Risk is calculated as long-term daily intake dose multiply cancer slope factor (SF), as a result of the incidence of cancer in exposed to carcinogens. The following formula were used:

$$\text{Low-dose exposure: } Risk = CDI \times SF_i \quad (Risk < 0.01). \quad (2)$$

$$\text{High-dose exposure: } Risk = 1 - \exp(-CDI \times SF_i) \quad (Risk > 0.01). \quad (3)$$

$$Risk(T) = \sum Risk_i. \quad (4)$$

$Risk_i$ : i Carcinogenic health risk;  $SF_i$ :  $\text{mg}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ ;  $CDI$ :  $\text{mg}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ .

Carcinogens are also non-carcinogenic risk. Non-carcinogenic risk (HI) is defined as the ratio between long-term daily intake dose and reference dose (U.S. EPA 1989):

$$H = CDI/RfD_i, \quad (5)$$

$RfD_i$  stands for Reference dose of pollutants,  $\text{mg}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ .

Reference dose of pollutants in evaluation are found in U.S. EPA (Table 1).

## RESULTS AND DISCUSSION

### CONCENTRATIONS OF PAHS IN THE ENVIRONMENT

The average concentrations of PAHs in water, soil and crops samples were  $8.5581 \mu\text{g}/\text{L}$ ,  $1.2507 \text{ mg}\cdot\text{kg}^{-1}$  and  $0.2543 \text{ mg}\cdot\text{kg}^{-1}$ , respectively. The results showed that the PAHs were prevalent in water, soil and crops. However, the distribution of PAHs in the study area was not uniform. For instance, there was clear spatial difference of PAHs in water samples (Figure 2). Compared with other regions in China, the concentration of PAHs is lower than the concentration of the Qiantang River in Hangzhou but higher than the concentration of the Yellow River Lanzhou section of Yangtze River, Songhua River, Wuhan section of Jiamusi section and the ring of the western region of Bohai (Bakar et al. 2014; Feng et al. 2011; Luo et al. 2005; Zuo et al. 2006). However, the concentration of PAHs recorded in the current study was higher than that report in Egypt, South Korea and Ghana (Amoako et al. 2011; Barakat et al. 2011; Kim et al. 2011; Maria et al. 2013).

TABLE 1. Chemical and toxicological properties of PAH<sub>16</sub>

PAH <sub>s</sub>	Nap	Ace	Flu	Ane	Phe	Ant	Fla	Pyr	Bgp
RfD	0.04	0.06	0.04	0.06	0.03	0.3	0.04	0.03	0.03
PAH <sub>s</sub>	Chr	Baa	Bbf	Bkf	Bap	Dbba	Ilp		
SF	0.0073	0.73	0.73	0.073	7.3	7.3	0.73		

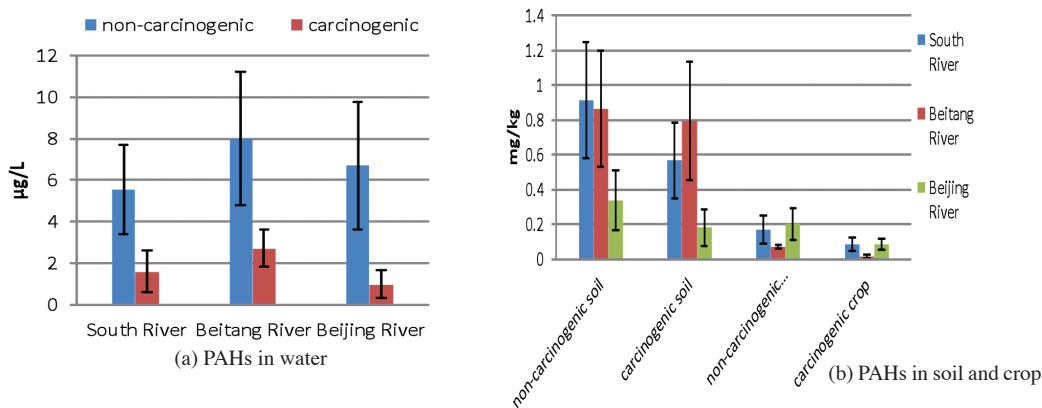


FIGURE 2. Residues of PAHs in three sewage irrigations

## EXPOSURE OF FARMERS IN SEWAGE IRRIGATION

Calculated according to the model, the total exposure of four age subsets (i.e. children, adolescents, adults and elderly) were 2.83, 2.34, 1.44 and 1.05  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , respectively. Weighted with the age span, daily life exposure is 1.59  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ . It was suggested that the weight difference was the main reason for exposure to decrease with age (Kopecka-Pilarczyk & Correia 2011). Excluding the impact of body weight, the absolute average daily exposure of children, adolescents, adults and the elderly were 51, 94, 101, 65  $\mu\text{g}\cdot\text{d}^{-1}$ , respectively. The absolute lifetime daily exposure weighted according to the age span is 86  $\mu\text{g}\cdot\text{d}^{-1}$ .

If we consider only nine carcinogenic PAHs (PAH<sub>9</sub>) (i.e. Nap, Ace, Flu, Ane, Phe, Ant, Fla, Pyr and Bgp), then weighted daily life exposure is 0.38  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , the total exposure of four age subsets were 0.78, 0.56, 0.33, 0.24  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , respectively. Excluding the impact of body weight, the absolute average daily exposure for children, adolescents, adults and the elderly were 14, 21, 23, 15  $\mu\text{g}\cdot\text{d}^{-1}$ , respectively and the absolute lifetime daily exposure PAH<sub>9</sub> weighted the age is 19  $\mu\text{g}\cdot\text{d}^{-1}$ . The exposure levels found in this study is higher than that reported before (i.e. children 8.6, adolescents 17.5, adults 20.4  $\mu\text{g}\cdot\text{d}^{-1}$ ) (Li et al. 2006). Compared with other parts of China, the exposure levels measured in this study is also higher than

the absolute lifetime daily exposure found in Beijing (63  $\mu\text{g}\cdot\text{d}^{-1}$ ), the exposure of children and adults in Lanzhou ( $0.428 \times 0.534 \mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ ) (Li et al. 2010, 2009). Compared with other countries, the exposure is far higher than the exposure of U.S.A (3.12  $\mu\text{g}\cdot\text{d}^{-1}$ ) (Gharibreza et al. 2013; Mitra & Bianchi 2003).

The absolute lifetime daily exposure in South River, Beitang River and Beijing River sewage irrigation areas were 1.85  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , 1.63  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$  and 1.29  $\mu\text{g}\cdot\text{kg}^{-1}\cdot\text{d}^{-1}$ , respectively (Figure 3).

As shown in Figure 4, the exposure levels in each age subset were different in different sewage irrigation areas. The proportions of exposure at different ages have some differences, followed by exposure risk size: children > adolescents > adults > the elderly. Children are the most sensitive age subset; they are at a critical period of growth and development, demand for various nutrients which are higher than adults. Regardless of the factor of weight, children even teenagers have higher exposures than adults under some exposure conditions.

Significant differences of PAHs exposure in three river irrigation areas were detected. The reason may lie in the difference in industrial structure and geographic characteristics (Aris et al. 2014; Lau et al. 2012). The order of exposure risk in three irrigation areas is South River sewage Irrigation > Beitang River sewage Irrigation > Beijing

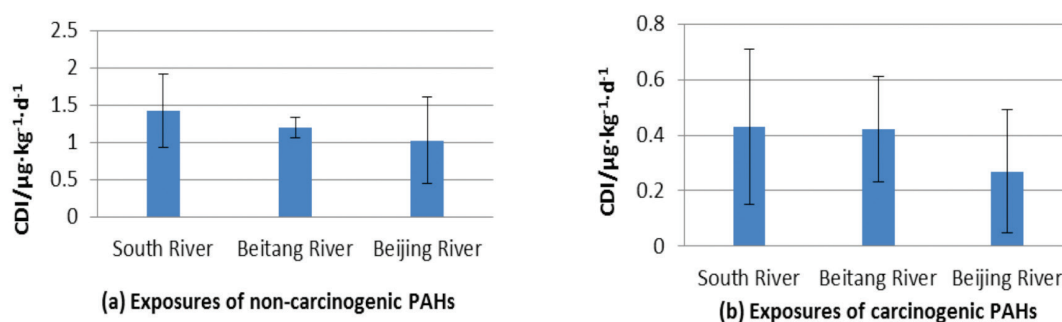


FIGURE 3. Exposures of PAHs

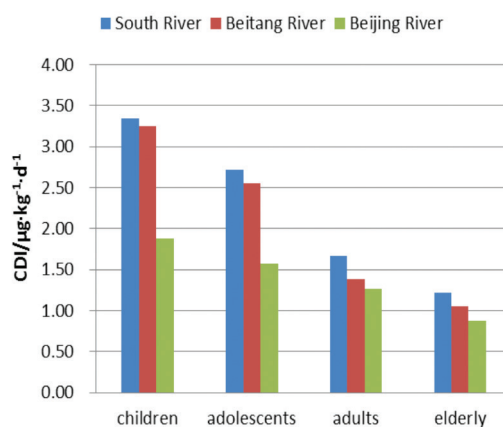


FIGURE 4. The exposure levels of four age groups in three sewage irrigations

TABLE 2. The health risks of PAHs

PAHs Risk	South River		Beitang River		Beijing River	
	non-carcinogenic	carcinogenic	non-carcinogenic	carcinogenic	non-carcinogenic	carcinogenic
Child	5.77E-03	5.40E-05	5.25E-03	6.24E-05	2.96E-03	3.87E-05
adole	4.72E-03	4.28E-05	4.22E-03	4.64E-05	3.17E-03	2.28E-05
Adult	2.91E-03	2.55E-05	2.35E-03	2.21E-05	2.32E-03	1.64E-05
Elderly	2.12E-03	1.88E-05	1.77E-03	1.82E-05	1.60E-03	1.14E-05

River sewage Irrigation. The value of South River sewage Irrigation is the highest. South River flows from west to south and there are many industries along the river which discharge large amounts of sewage. For example, a large chemical plant near Yutai Village discharge large amount of wastewater every week with multiple colour. In the area at south of Haihe River, the amount of domestic wastewater discharge each year is considerable. Therefore, South River sewage irrigation pollution contains two parts: Industrial wastewater and municipal sewage. Beitang River is the shortest river of the three study areas. It flows from the east and its main receiving water body comes from north of the Haihe municipal sewage and smelting, chemical and other industrial wastewater. Beijing River runs via northern of Tianjin to Bohai Sea. It is an important branch of the Beijing municipal wastewater discharge, receiving Beijing's main industrial wastewater and domestic sewage in rural areas of Tianjin.

#### HEALTH RISK ASSESSMENT

The personal health risks caused by PAHs in three study areas were calculated using the health risk assessment models (Table 2).

According to U.S. EPA definitions, the pollutant would cause harm to human health when the non-carcinogenic risk value over 1 (U.S. EPA 1992). The statistical analysis showed that non-cancer risk for sewage irrigation is  $2.72 \times 10^{-3}$ , non-carcinogenic exposure found in the three study areas is in the following order: South River ( $3.24 \times 10^{-3}$ ) > Beitang River ( $2.74 \times 10^{-3}$ ) > Beijing River ( $2.33 \times 10^{-3}$ ). The results indicated that contaminants in three study areas did not have a significant non-carcinogenic health hazard to the population.

For carcinogenic risk, U.S EPA recommended the acceptable risk value is  $1.0 \times 10^{-4}$ . U.S EPA recommended that in general the acceptable cancer risk is  $10^{-6}$ . The results in this study showed that the carcinogenic risk of sewage irrigation is  $2.49 \times 10^{-5}$ , the carcinogenic risk in three study areas is South River ( $2.89 \times 10^{-5}$ ) > Beitang River ( $2.83 \times 10^{-5}$ ) > Beijing River ( $1.79 \times 10^{-5}$ ). Compared with the previous reports, carcinogenic risk of sewage irrigation in Tianjin is lower than that in Beijing ( $3.1 \times 10^{-5}$ ), in Jiamusi section of Songhua River ( $6.52 \times 10^{-5}$ ), and the Lanzhou section of the Yellow River ( $4.46 \times 10^{-5}$ ), but higher than that in Ningbo ( $3.17 \times 10^{-7}$ ) (Feng et al. 2011; Luo et al. 2005; Sun et al. 2012; Wang et al. 2011; Yusoff et al. 2013; Zuo et al. 2006).

#### CONCLUSION

Because of applied EPA parameters, there were limitations on the evaluation of Chinese population health risks. Although the risk of contaminants still within the acceptable range, from the perspective of long-term security, carcinogenic health hazards arising crowd has reached a high level. In particular, PAHs pose serious health risks to children. Therefore, the sewage irrigation areas in Tianjin have serious health risks according to the results obtained in this study. Hence, we suggest that crops of sewage irrigated areas and their distribution channels should be re-concerned by the government in the future city planning.

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