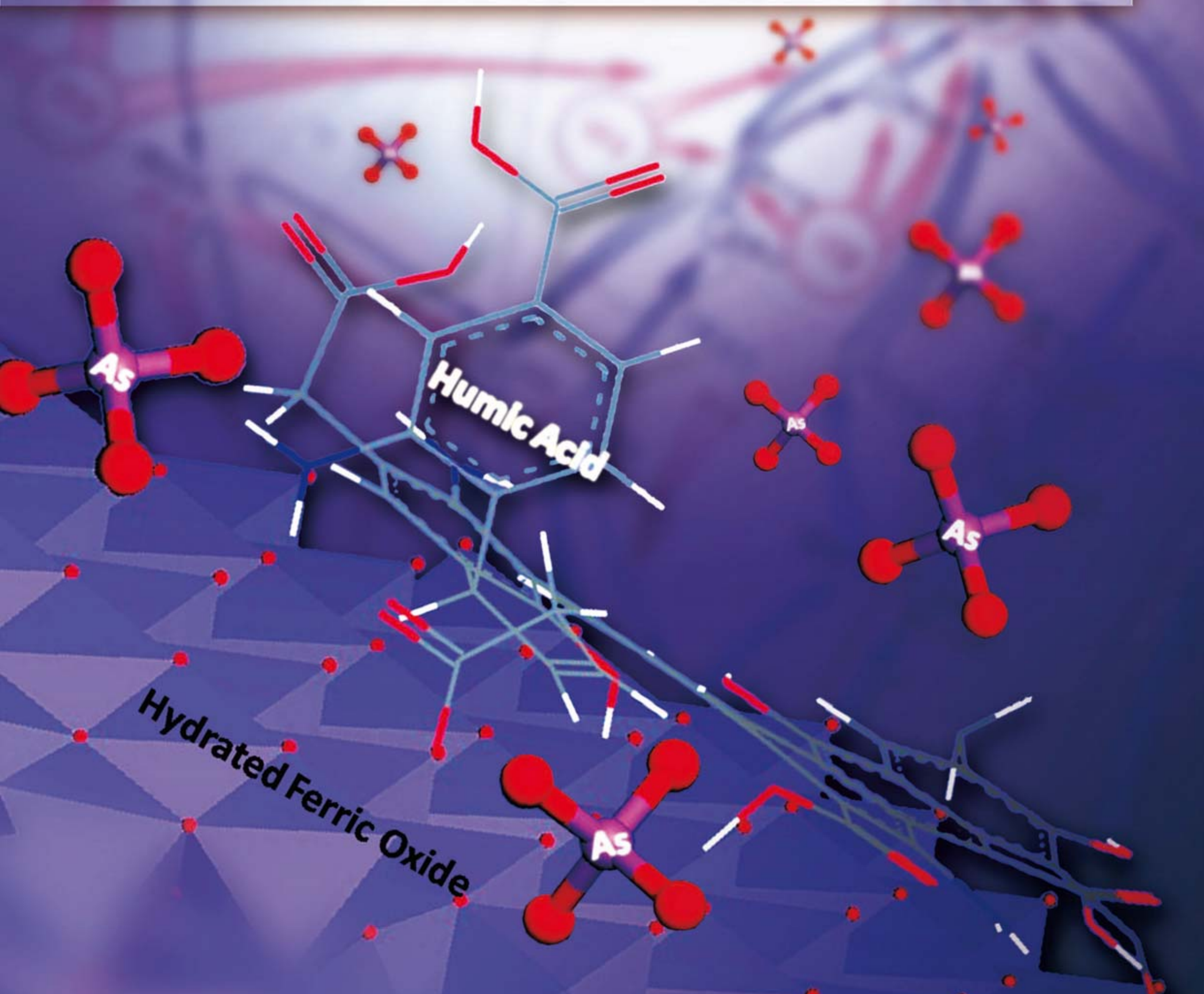
JES

JOURNAL OF ENVIRONMENTAL SCIENCES

ISSN 1001-0742
CN 11-2629/X

February 1, 2014 Volume 26 Number 2
www.jesc.ac.cn



ELSEVIER
Science Press

Sponsored by
Research Center for Eco-Environmental Sciences
Chinese Academy of Sciences

ISSN 1001–0742 **Journal of Environmental Sciences** Vol. 26 No. 2 2014

CONTENTS

Environmental catalysis and materials

Serial parameter: CN 11-2629/X*1989*m*261*en*P*30*2014-2

Journal of Environmental Sciences 26 (2014) 432–439

Available online at www.sciencedirect.com

Journal of Environmental Sciences

www.jesc.ac.cn

Health risk assessment of dietary exposure to polycyclic aromatic hydrocarbons in Taiyuan, China

Jing Nie[1,2,**], Jing Shi[3,**], Xiaoli Duan[1,*], Beibei Wang[1], Nan Huang[1], Xiuge Zhao[1]

1. Chinese Research Academy of Environmental Sciences, State Key Lab of Environmental Criteria and Risk Assessment, Beijing 100012, China. E-mail: niejing@craes.org.cn (Jing Nie); shijing8008@163.com (Jing Shi)

2. Soil and Water Science, College of Resources and Environmental Sciences, China Agricultural University, Beijing 100094, China

3. Department of Safety Engineering, China Institute of Industrial Relations, Beijing 100048, China

ARTICLE INFO

Article history:
Received 11 March 2013
revised 11 April 2013
accepted 20 June 2013

Keywords:
dietary exposure;
food;
health risk assessment;
polycyclic aromatic hydrocarbons (PAHs);
benzo[a]pyrene

DOI: 10.1016/S1001-0742(13)60424-6

ABSTRACT

Sixteen polycyclic aromatic hydrocarbons (PAHs) were determined in 24 duplicate-diet samples from people in Taiyuan during summer and winter in 2009. Dietary intake of PAHs for 2862 participants was subsequently estimated by a survey in Taiyuan. Results from these 24 samples were compared with a raw food study in Taiyuan in 2008. Three main sources of dietary PAHs are vegetables, wheat flour and fruits, the sum of which contributes 75.95% of PAHs in dietary food. Compared to the estimated value in raw food, much more B[a]P_{eq} (benzo[a]pyrene equivalents) were detected in food samples collected in the duplicate-diet study in Taiyuan (60.75 ng/day). The cooking process may introduce more B[a]P_{eq} into food, and the relative contribution of 16 PAHs in the diet would be changed during the cooking procedure.

Introduction

Polycyclic aromatic hydrocarbons or polynuclear aromatic hydrocarbons (PAHs) are potent atmospheric pollutants that consist of fused aromatic rings and do not contain heteroatoms or carry substituents. They are widely believed to make a significant contribution to human cancer risk. The sources of PAHs are well known as forest fires, vehicular emissions, residential wood burning and industrial combustion of fossil fuels. These environmental pollutants subsequently are found in air, water, soil and food stuffs. The US Environmental Protection Agency (EPA) has classified benzo[a]anthracene, benzo[a]pyrene (B[a]P), benzo[b]fluoranthene, chrysene, benzo[k]fluoranthene, dibenzo[a,h]anthracene, and indeno[1,2,3-c,d]pyrene as probable human carcinogens (group B2) (US EPA, 2002). The International Agency for Research on Cancer (IARC) established that benzo[a]anthracene and benzo[a]pyrene are probable human carcinogens, whereas benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-c,d]pyrene are possible human carcinogens (IARC, 2004).

Some researchers suggest that there are different levels of PAHs in daily food consumption, which may induce more health effects on human beings (Voutsa and Samara, 1998; Li et al., 2005; Yoon et al., 2007; Martí-Cid et al., 2008; Perelló et al., 2009; Zhang et al., 2010). It is widely reported that the PAHs level varied significantly with different food types (Grung et al., 2009; Ciemniak

* Corresponding author. E-mail: duanxiaoli@263.net;

** The authors contribute equally to this work.

and Chrachol, 2008; Cejpek et al., 1998; Kuhn et al., 2009; Duedahl-Olesen et al., 2011; Hossain and Hoque, 2011; Levengood and Schaeffer, 2011; Santos et al., 2011). Several studies estimated daily PAHs or B[a]P equivalent (B[a]P_{eq}) intake levels based on PAH levels in raw food. Xia et al. (2011) suggested an estimated median value of B[a]P_{eq} daily exposure for adults was 0.57 μg/day, based on PAH concentrations of 25 kinds of raw food in Taiyuan, China, while Li (2007) estimated 13.72 to 40.74 μg/day for the general population in Tianjin, China. In Spain, the estimated total PAH levels were 6.71 μg/day for male adults and 3.92 μg/day for female adults (Martí-Cid et al., 2008). A study of the Dutch diet estimated an average PAH daily intake of 5–17 μg/day (de Vos et al., 1990). In New Zealand, the mean dietary intake level of PAHs was estimated at 1.2 μg/day (Thomson et al., 1996).

However, few studies have been conducted on the PAHs from cooked food consumption. It is widely known that PAHs in food might rapidly increase during cooking. It is more reasonable to use PAHs in cooked food to reflect PAH exposure level in residents. This research focuses on the PAH contamination level of duplicate-diet samples in Taiyuan, a region known in China as 'highly' contaminated by the coal industry in terms of coal burning smoke and vehicular emissions. Duplicate-diet (duplicate plate) samples were used, i.e., subjects were asked to prepare two identical plates for each meal, one for actual consumption, and the other for sample collection. Our main objective was to determine the PAH levels in cooked or consumed food, the major sources of exposure/contamination by PAHs, and the cancer risk of cooked food in Taiyuan.

1 Experimental

1.1 Sampling

Taiyuan City, Shanxi Province, is one of the most polluted cities in China because of coal burning by industry in combination with vehicle emissions, and its typically low wind speed (2.4–2.5 m/sec, annual average) cannot diffuse the pollutants. PAHs may thus be a threat to the health of local residents (Xia et al., 2011). The study was carried out in 24 adult subjects, including 18 males and 6 females with 9 of them from rural areas, 10 from urban areas with non-occupational exposure and 5 from the industrial areas. Then 2850 adult subjects were selected to participate in a survey to study the food consumption habits and food consumption amounts by questionnaire. The subjects chosen are consistent with the population distribution of Taiyuan. The sort of food in the survey includes fish, chicken, pork, beef, mutton, vegetables, fruits, milk, eggs, rice, flour and edible oil, the cooking types, eating frequency and account of which were all investigated. The survey was accomplished through face to face household survey methods with 10% parallel questionnaires to control quality. The questionnaires and parallel questionnaires were entered simultaneously in EpiData, and differences of before and after input rate were less than 3%.

1.2 Analysis

All food samples within a 24-hr study period were collected using the 'duplicate plate' method. Each subject was provided with 4 Tupperware containers and 1 cooler with ice packs for the collection of meal samples. These samples were kept in a cooler with icepacks until they were returned to the laboratory. After being weighed, all the samples within the 24-hr period were mixed evenly to form a composite sample for each subject. A form was administered to each subject to record their sampling time, kinds of food with the cooking type, like fried chicken, and laboratory coding during the sampling.

Food samples were prepared using the following procedure. An aliquot (100 g) of each composite sample was placed into a 1-L Erlenmeyer flask containing 12.5 g KOH (Beijing Chemical Reagent, Beijing, China) in 200 mL of 95% ethanol (Beijing Chemical Reagent, Beijing, China) and a mixing stirrer. After being treated at 80°C for 4 hr, the entire content of the flask was filtered through a filter paper (0.45 μm); and the filtrate was transferred into a 2-L separatory funnel. Liquid-liquid extraction was performed using 60 mL isooctane (Beijing Chemical Reagent, Beijing, China) for 10 min and then repeated to separate PAHs from ethanol (Beijing Chemical Reagent, Beijing, China). The isooctane phase was transferred to a 350 mL amber round flask and concentrated to 5 mL using a vacuum rotary evaporator (R-201, Shanghai, China) at 60°C. If visible solid phase still was present in the condensed extract, centrifugation was used to remove the solid residues. Sample enrichment and purification were completed using a Sep-Pak Florisil cartridge (Waters051960, Waters Corp., USA). The cartridge was first primed with 5 mL isooctane. After the condensed sample extract passed through the cartridge at a flow rate of 1.0 mL/min, 10 mL benzene was introduced into the cartridge to extract PAHs from the cartridge coating material. The benzene solution in the eluent was evaporated to 0.1 mL using a nitrogen gas flow. Acetonitrile was added to make a 1 mL final solution that was further filtered through a 0.2 μm PVDF Liquid Filter into an auto-sampler amber vial sealed with a Teflon septum. The vial was wrapped with aluminum foil and stored in a freezer at $\leqslant$ –7°C before GC-MS analysis. All samples were protected from direct exposure to light during sample preparation. Quantitative analysis of the food sample extracts was done by gas chromatography with mass spectrometer detector (GC6890/MS5975, Agilent, USA). A 30 m × 0.25 mm i.d. × 0.25 μm film thickness DM-5MS/LB capillary column (Agilent, USA) was used, and the temperature was programmed from an initial 60°C to 150°C at 20°C/min with a holding time of 1 min; secondly up to 230°C/min at 6°C/min with a holding time of 1 min; thirdly up to 240°C/min at 1°C/min with a holding time of 2 min; finally up to 300°C/min at 8°C/min with a holding time of 10 min. A 1.0 μL aliquot of the extract was injected while Helium was used as the carrier gas and the injector port was operated at a flow rate of 1.0 mL/min, with Gasification chamber temperature: 280°C; Interface temperature: 290°C; Ion source temperature: 230°C; and Level quadrupole temperature: 150°C.

1.3 Data analysis

All the statistical analyses were conducted by the software SAS 9.2.1 (SAS institute, Cary, NC, 2008). The sample recoveries are between 60% and 120%. A parallel blank spiked sample was run every 10 samples during the sample pre-treatment process and a standard sample was run every 10 samples during the analysis of the test. The test was applied to study the equality of variances. The statistical significance of differences was assessed by applying the Kruskal-Wallis test because data did not meet the equal variance assumption. A probability of 0.05 or lower ($p \leqslant 0.05$) was considered to be significant.

1.4 Dietary exposure and cancer risk assessment

The equivalents of the mixture were multiplied by the potency for B[a]P (US EPA, 2002; Yoon et al., 2007). The toxic equivalency factors (TEFs) adopted by the US EPA (2002) were 0.001 for benzo[a]anthracene, acenaphthene, acenaphthylene, fluorine, phenanthrene, fluoranthene, and pyrene; 0.01 for anthracene, chrysene, and benzo[g,h,i]perylene; 0.1 for benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, and indeno[1,2,3-c,d]pyrene; and 1 for benzo[a]fluoranthene and dibenzo[a,h]anthracene. TEF values were used to calculate PAHs as benzo[a]pyrene equivalents for a male adult resident in Taiyuan. This estimates the potency of B[a]P and expresses the environmental levels of other PAH as 'B[a]P equivalents, B[a]P_{eq}'.

Total dietary intake of PAHs was calculated by summing the results of multiplying each dietary food by the consumed amount, which was calculated from the questionnaire results. The carcinogenic potencies of 16 PAHs were estimated as the sum of each individual B[a]P_{eq}. The TEFs from Petry et al. (1996) were adopted to calculate B[a]P_{eq}. The incremental lifetime cancer risk (ILCR) of population groups in Taiyuan caused by PAHs dietary exposure was calculated by the following equation.

$$\text{ILCR} = E_{\text{D}} \times \text{EF} \times \text{ED} \times \text{SF} \times \text{CF}/(\text{BW} \times \text{AT}) \quad (1)$$

where the ILCR refers to the incremental lifetime cancer risk of the dietary exposure, E_{D} (ng/day) is the daily dietary PAH exposure level, ED (yr) is the exposure duration (for adults: ED=43 yr), EF (365 days/yr) is the exposure frequency, SF is the oral cancer slope factor of benzo[a]pyrene (4.5, 5.9, 9.0 and 11.7, with a geometric mean of 7.3 (mg/(kg·day)) (Brune et al., 1981; US EPA 1991a, 1991b, 2001), CF (10^{-6} mg/ng) refers to conversion factor, BW (kg) is the body weight, AT (25,550 days) is the average lifespan for carcinogens.

2 Results and discussion

2.1 Results

Sixteen PAH concentrations in dietary food were detected (**Table 1**). Phenanthrene, fluoranthene, pyrene were found to be higher than the other 13 PAHs in the dietary food samples. PAHs varied greatly in different diets. According to the consumption food categories, the highest total PAHs was in food from farmers, and residents in the downtown city (10.22 ng/g) dominated by rice, potatoes and pork. Compared to this study, the estimated total PAH concentrations from raw food are a little higher, especially for male adults. Daily PAHs in daily consumed food were estimated by raw food composition. Rice, wheat flour, beef and mutton, pork, chicken, vegetables, fruits, milk and oils were assumed to be consumed every day, while in our study, the major categories of food consumption were mainly rice, wheat flour, vegetables and eggs for Taiyuan residents. Some of the subjects (62.50%) consumed only wheat and vegetables all day during the sampling period. The level of total PAH intake by food might be higher than for uncooked foods, and levels of PAHs might be greatly changed during heating and frying.

B[a]P in daily consumed food was much higher in cooked food than values estimated from raw food. Especially for female adults, B[a]P increased to as much as 33 times higher than that in raw food. Some studies show that PAHs are formed when food is cooked over an open flame, however normal roasting or frying food does not produce, or limitedly produces copious quantities of PAHs (Perelló et al., 2009; Howard and Fazio, 1980). In this study, much higher B[a]P was detected in cooked food, which was just cooked with normal methods. This may because the oils are always heated at high temperatures during Chinese food cooking. The high incidence of lung cancer observed among Chinese women has been shown to be associated with exposure to fumes from cooking oil, which emits PAHs when the oils are heated at high temperatures (Pan et al., 2008). Finally, concentrations of PAHs in duplicate-diet samples in this study are higher than those in other countries. This result was similar to some other studies (Li et al., 2005; Xia et al., 2011).

The amount of daily food consumption in this study, the Health and Diet survey in 2002 in Taiyuan and United States data in the US EPA exposure factor handbook are shown in **Table 2**. The amount of food consumption in this study was estimated by questionnaires with 2862

Table 1 Comparison of duplicate-diet PAH concentrations (ng/g) in 24 hr solid food of Taiyuan participants and surveys of other countries

PAHs	PAHs in this study in Taiyuan			PAHs in raw food in Taiyuan*			PAHs in raw food in other research						
	Male and female	Male	Female	Male and female	Male	Female	Anshan, China, 2008[a*]	Qujing, China, 2010[b*]	Tianjin, China, 2007[c]	Spain, 2008[d]	USA, 2001[e]	Dutch, 1990[f]	United Kingdom, 1983[g]
Naphthalene	5.28	4.58	7.37	44.26	48.52	39.99	8.30	20.70	30.8–72.7	0.91	2.16	–	–
Acenaphthene	1.42	1.39	1.49	2.86	3.12	2.60	1.10	1.30	1.0–1.7	0.40	–	–	–
Acenaphthylene	1.47	1.52	1.30	1.53	1.66	1.39	–**	1.30	1.6–3.3	0.16	–	–	–
Fluorene	6.54	6.20	7.54	7.27	7.90	6.64	3.00	4.10	4.1–7.7	0.39	–	–	–
Phenanthrene	12.20	10.45	17.43	21.32	23.17	19.47	9.30	6.30	8.9–16.8	2.31	–	4.51	–
Anthracene	1.99	1.67	2.93	2.38	2.58	2.17	1.50	0.90	1.8–3.4	0.58	–	0.64	0.99
Fluoranthene	4.68	2.62	10.84	6.60	7.15	6.05	19.00	8.20	3.2–6.0	0.83	4.41	1.66	–
Pyrene	3.97	2.35	8.84	4.53	4.91	4.15	3.60	11.30	1.4–2.3	0.95	1.44	–	–
Benzo[a]anthracene	1.69	0.79	4.38	0.62	0.68	0.56	1.00	2.20	0.1–0.3	0.13	–	0.36	0.22
Chrysene	2.19	1.18	5.20	0.61	0.66	0.56	1.10	2.00	0.8–1.6	0.13	–	1.53	0.50
Benzo[b]fluoranthene	0.95	0.60	2.00	0.58	0.63	0.53	0.50	1.00	2.7–4.4	0.09	–	0.36	0.18
Benzo[k]fluoranthene	0.31	0.14	0.81	0.18	0.19	0.16	0.20	0.20	0.5–0.9	0.06	0.26	0.14	0.06
Benzo[a]pyrene	1.30	0.51	3.68	0.12	0.13	0.11	0.50	0.30	0.8–2.0	0.08	0.48	0.29	0.25
Dibenzo[a,h]anthracene	0.20	0.09	0.53	0.02	0.02	0.02	n.d.	n.d.	0.1–0.2	0.05	–	–	0.33
Indeno[1,2,3-c,d]pyrene	2.02	1.61	3.26	0.10	0.11	0.09	–	n.d.	2.5–7.6	0.07	–	–	–
Benzo[g,h,i]perylene	1.83	1.35	3.27	0.16	0.17	0.14	0.60	n.d.	0.2–0.4	0.05	0.81	0.36	
Total PAH	48.01	37.06	80.86	93.11	101.61	84.61	49.70	59.80	60.5–131.4	7.19	9.56	9.85	2.53

* Data are shown as median value. ** Not determined. [a] Sha, 2008; [b] Zhang, 2010; [c] Li, 2007; [d] Martí-Cid et al., 2008; [e] Kazerouni et al., 2001; [f] de Vos et al., 1990; [g] Dennis et al., 1983.

Table 2 Comparison of daily food consumption (g/day) for male and female adults in this study, the general population of Taiyuan and USA

Food	This study			General population in Taiyuan*			General population in USA**
	Male and female	Male	Female	Male and female	Male	Female	Male and female
Rice	66.25	77.5	56.75	71.48	79.03	63.92	24.00
Wheat flour	371.25	441.25	306.25	260.91	286.52	235.30	186.00
Vegetables	430	453.75	410	165.14	172.99	157.29	309.00
Fruits	129.64	132.78	126.89	18.04	15.13	20.94	191.00
Beef and mutton	6.30	7.14	5.41	8.07	9.24	6.90	70.00
Chicken	8.83	9.92	7.88	2.33	2.57	2.09	63.00
Pork	41.5	46.75	36.25	19.7	21.77	17.63	32.90
Fish	9.71	10.75	8.71	3.89	4.21	3.57	15.00
Eggs	35	36.75	33.5	12.61	12.87	12.34	23.10
Milk	41.47	40.94	41.9	17.06	16.30	17.81	231.00
Oils	102.04	102.04	102.04	25.96	27.47	24.45	64.00
Total	1241.99	1359.57	1135.58	648.10	562.24	605.17	1209.00

* Ingestion amounts of foods are obtained from the data of 2002 Health and Diet Survey in Taiyuan, China, using interpolation method for some missing data (Ge, 1992; Wang, 2005; Zhai and Yang, 2006) and the data are presented as means.

** Data are obtained from the US EPA exposure factor handbook (US EPA, 2009).

subjects in Taiyuan City. The highest quantity food types consumed are vegetables, followed by wheat flour, fruits and oils. Compared with data from 2002, people consume much more wheat flour products, vegetables and fruits in Taiyuan. This might be due to the increase of food supplements and food composites in China. People in the United States consume more fruits, milk and meat per day than those in our research.

In the questionnaires, daily oils consumption for the general population of Taiyuan is 102.04 g/day, which is higher than the data of the 2002 Health and Diet Survey in Taiyuan (25.96 g/day) and general population in the USA (64.00 g/day). This might be due to the improvement in people's living standards and differences in food cooking habits between Chinese and American people. Taiyuan people consume much more fried food than people in the United States.

The contribution of each food category to the total PAH levels is depicted in **Fig. 1**. Vegetables (34.62%) contributed more than one-third of the total, followed by wheat flour (29.89%) and fruits (10.44%). According to the results, the specific contributions of each food category have changed compared to those observed in raw food. Some studies indicated that PAHs in food would be higher after cooking (Perelló et al., 2009; Howard and Fazio, 1980; Ward et al., 1997). But other studies showed

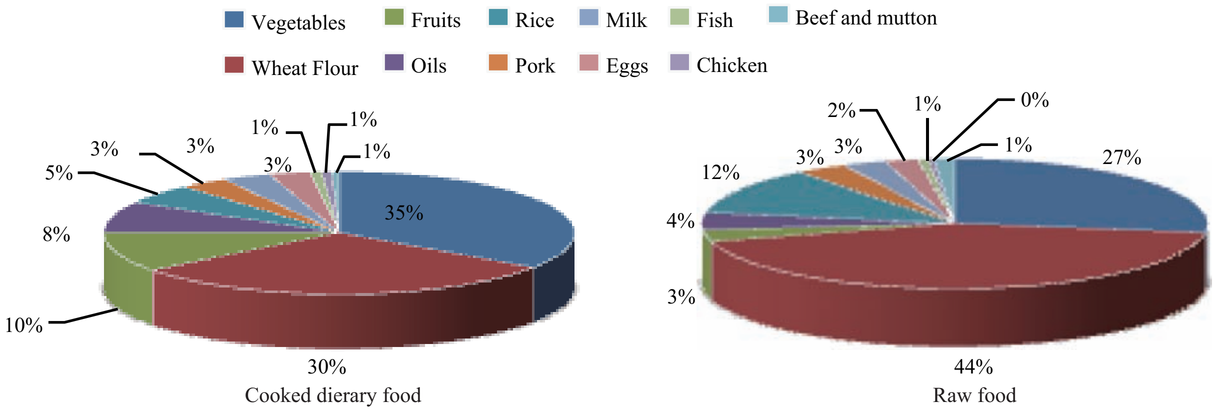


Fig. 1 Percentage contributions to B[a]P equivalents by various foods.

inverse results, that normal roasting or frying food does not produce copious quantities of PAHs (Howard and Fazio, 1980). Our study indicates that the contributions to total dietary intake of PAHs or B[a]P_{eq} by different kinds of food would change after cooking, and the PAH concentration varies for different food categories after cooking.

Results of PAHs from dietary intake by residents in Taiyuan and other countries are shown in **Table 3**. Total PAHs are at a similar level between our studies from cooked food and those from raw food, while much higher naphthalene was estimated in raw dietary food. Dietary intake of total PAHs is the highest (60.75 ng/day) in our study, compared with surveys in other countries. Females consumed much more PAHs than males did, which is different from values estimated from raw food. This could be explained by the difference in food habits between genders in Taiyuan. In this study, female participants consumed more vegetables, wheat flour and fruits, which may contribute relatively higher amounts of PAHs, while male participants usually consumed more meat, oils and fats than females. That can explain the results estimated from raw food. But male participants consumed less meat products relative to vegetables and fruits.

Mean dietary intake of B[a]P or B[a]P_{eq} in China and other counties is shown in **Table 4**. The highest B[a]P_{eq} intake was detected in populations in Tianjin, which included more categories of food and beverage (Li et al., 2005; Li 2007). This may due to the much higher dietary exposure dose in both vegetables and oils, as well as the ingestion amount of some foods in Tianjin being significantly higher than that in Taiyuan. The B[a]P_{eq} level in this study is higher than other studies except Tianjin. Vegetables contribute the most B[a]P_{eq} (4720.69 ng/day), followed by fruits (1423.23 ng/day) and oils (1120.23 ng/day). In

Table 3 Comparison of dietary intakes of PAHs (ng/day) by adults living in Taiyuan and other countries

PAHs	Dietary intakes of PAHs in this study in Taiyuan*			Dietary intakes of PAHs by raw food in Taiyuan*			Dietary intakes of PAHs from various surveys			
	Male and female	Male	Female	Male and female	Male	Female	United Kingdom, 2000 [a]	Italy, 1993–1995 [b]	Austria, 1989 [c]	The Netherlands 1984–1986 [d]
Naphthalene	6.77	6.23	8.37	28.68	31.44	25.92	0.25	–	–	–
Acenaphthene	1.84	1.89	1.69	1.85	2.02	1.68	–**	–	–	–
Acenapthylene	1.92	2.07	1.48	0.99	1.08	0.90	–	–	–	–
Fluorine	8.46	8.43	8.56	4.71	5.12	4.30	–	–	–	–
Phenanthrene	15.61	14.21	19.79	8.21	15.02	1.40	2.73	–	<0.33	5.13
Anthracene	2.54	2.27	3.33	1.92	1.67	2.17	0.14	–	<0.04	0.70
Fluoranthene	5.75	3.56	12.31	4.28	4.63	3.92	0.60	–	0.60	2.11
Pyrene	4.90	3.19	10.04	2.94	3.18	2.69	0.60	–	0.60	–
Benzo[a]anthracene	2.05	1.07	4.97	0.40	0.44	0.36	0.10	0.41	<0.02	0.65
Chrysene	2.68	1.60	5.91	0.40	0.43	0.36	0.19	1.46	0.20	3.90
Benzo[b]fluoranthene	1.18	0.82	2.27	0.38	0.41	0.34	0.18	–	0.01	0.59
Benzo[k]fluoranthene	0.37	0.19	0.92	0.12	0.13	0.11	0.15	–	0.04	–
Benzo[a]pyrene	1.56	0.69	4.18	0.08	0.09	0.07	0.19	0.17	0.05	0.42
Dibenzo[a,h]anthracene	0.24	0.12	0.60	0.01	0.01	0.01	0.06	0.08	<0.02	–
Indo[1,2,3-c,d]pyrene	2.57	2.19	3.70	0.07	0.07	0.06	0.17	0.16	<0.02	0.55
Benzo[g,h,i]perylene	2.31	1.84	3.71	0.10	0.11	0.09	0.11	–	0.12	0.20
Total PAH	60.75	50.39	91.82	60.35	65.85	54.84	5.47	2.28	<1.62	14.25

* Data was calculated from questionnaire survey results in Taiyuan.

** Not determined.

[a] UK FSA_0, 2002; [b] Lodovici et al., 1995; [c] Pfannhauser, 1991; [d] de Vos et al., 1990.

Table 4 Mean dietary intake (ng/day) of B[a]P or B[a]P equivalents in China and other countries*

Country and cities	Years of publication	Total intake	Vegetables	Fruits	Meat			Fish	Eggs	Milk	Oils	Reference
					Pork	Chicken	Mutton and beef					
Taiyuan, China	–	786 [a], 13635 [b]	272.13 [a], 4720.69 [b]	82.04 [a], 1423.23 [b]	26.26[a], 455.60 [b]	5.59 [a], 96.94 [b]	3.99 [a], 69.16 [b]	6.15 [a], 106.6 [b]	22.15 [a], 384.24 [b]	26.24 [a], 455.27 [b]	64.58 [a], 1120.23 [b]	This study
Taiyuan, China [b]	2011	3543–8140 [b]	22.52 [b]	5.97 [b]	33.23 [b]	1.49 [b]	24.61 [b]	22.23 [b]	3.25 [b]	3.11 [b]	17.91 [b]	Xia et al., 2011
Tianjin, China	2007	3543–8140 [b]	520.19 [b]	3.97 [b]	93.18 [b]	11.02 [b]	38.17 [b]	5.91 [b]	3.91 [b]	4.09 [b]	122.53 [b]	Li, 2007
Catalonia, Spain [b]	2008	760 [b]	12.32 [b]	4.00 [b]	10.65 [b**]			4.00 [b]	2.56 [b]	2.00 [b]	12.56 [b]	Marti-Cid et al., 2008
Korea	2007	162.06	51.19	0.90	8.67	1.84	4.92	1.21	6.68	3.58	n.d.	Yoon et al., 2007
Greece	1998	–	4.82	–**	–	–	–	–	–	–	–	Voutsa and Samara, 1998

* Data are obtained by simple calculation. n.d.: not determined.

** Not determined.

[a] Data are given as B[a]P.

[b] Data are given as B[a]P equivalents.

** Including all meat and 7 kinds of meat and 4 kinds of meat products in Catalonia.

Table 5 Dietary incremental lifetime cancer risk for population in Taiyuan

PAH	Intake (ng/day)[a]	TEF (US EPA, 2002)	Cooked food			Raw food		
			Intake of Ba[a]P equivalents (ng/day)	B[a]P (%)[b]	Lifetime cancer risk ($\times10^{-6}$)	Intake of B[a]P equivalents (ng/day)	B[a])P (%)	Lifetime cancer risk ($\times10^{-6}$)
Benzo[a]anthracene	6.77	0.001	0.01	0.27	2.16	0.03	10.95	12.22
Acenaphthene	1.84	0.001	0.00	0.07	0.54	0.00	0.71	0.79
Acenaphthylene	1.92	0.001	0.00	0.08	0.57	0.00	0.38	0.42
Fluorene	8.46	0.001	0.01	0.33	2.59	0.00	1.80	2.01
Phenanthrene	15.61	0.001	0.02	0.61	4.94	0.01	3.14	3.50
Anthracene	2.54	0.01	0.03	1.00	7.96	0.02	7.33	8.18
Fluoranthene	5.75	0.001	0.01	0.23	1.99	0.00	1.63	1.82
Pyrene	4.90	0.001	0.00	0.19	1.69	0.00	1.12	1.25
Benzo[a]anthracene	2.05	0.1	0.20	8.05	72.72	0.04	15.28	17.04
Chrysene	2.68	0.01	0.03	1.05	9.40	0.00	1.51	1.68
Benzo[b]fluoranthene	1.18	0.1	0.12	4.66	40.41	0.04	14.32	15.97
Benzo[k]fluoranthene	0.37	0.1	0.04	1.47	13.36	0.01	4.58	5.11
Benzo[a]pyrene	1.56	1	1.56	61.52	570.44	0.08	30.56	34.08
Dibenzo[a,h]anthracene	0.24	1	0.24	9.45	86.44	0.01	3.82	4.26
Indeno[1,2,3-c,d]pyrene	2.57	0.1	0.26	10.11	84.15	0.01	2.48	2.77
Benzo[g,h,i]perylene	2.31	0.01	0.02	0.91	7.58	0.00	0.38	0.43
Total	60.75	–	2.54	100.00	906.95	0.26	100.00	111.51

[a] Food intake amounts were calculated as 1.36 ng/day and 0.94 ng/day as evaluated in Sun's study.

[b] Percentages of contributions to the B[a]P equivalents. The B[a]P equivalents are the sums of the results of multiplying the intake of each of the respective seven or eight PAH by their respective TEFs.

Catalonia, B[a]P_{eq} in each kind of food was much lower than that in Taiyuan, and oils and vegetables contributed more to the total contamination. Vegetables were also the biggest contributor to the daily B[a]P_{eq} consumption in Korea.

2.2 Cancer risks of dietary PAH exposure

According to the US EPA, a one in a million chance of additional human cancer over a 70 year lifetime (ILCR=10^{-6}) is the level of risk considered acceptable or inconsequential; whereas additional lifetime cancer risk of one in ten thousand or greater (ILCR=10^{-4}) is considered serious. There is high priority for paying attention to such health problems. **Table 5** shows dietary incremental lifetime cancer risk for the population in Taiyuan. In this study, the mean value of ILCRs for subjects in Taiyuan is higher than the priority risk level (10^{-4}). In contrast, for the raw food, the median values of ILRS for all population groups fell within the range of 10^{-6}–10^{-5}, being higher than the acceptable risk level (10^{-6}) and lower than the priority risk level (Xia et al., 2011). For daily consumed food, the highest contribution corresponded mainly to benzo[a]pyrene (61.52%). This is in agreement with the results in the raw food. Indeno[1,2,3-c,d]pyrene has the second highest contribution in daily consumed food (10.11%), while benzo[a]anthracene is the second highest contributor to lifetime cancer risk (17.04×10^{-6}). This indicates that the dietary food consumption which includes cooked food and raw food would pose much more cancer risk than raw food itself.

3 Conclusions

Results indicate that the main sources of dietary PAHs in Taiyuan residents are vegetables, wheat flour and fruits, the sum of which contributes 75.95% of PAHs in dietary food. One of the important reasons for this is that people in Taiyuan consume much more wheat flour products than in any other region in China.

In this study, lower PAH concentrations and much higher B[a]P_{eq} daily consumption were found in duplicate-diet food samples. This may be because the types of food consumed were relatively few. Participants consumed more cereals and vegetables but less meat in the sample collection period. It also suggests that cooking may induce the increase of B[a]P_{eq}.

Compared with those in other cities of China and other countries, an excess risk of cancer for residents in Taiyuan was observed. Compared to raw food, cooked or consumed food induces much higher cancer risks.

The contributions of 16 PAHs to food intake contamination would be changed after cooking. For daily consumed food, the highest contribution corresponded mainly to benzo[a]pyrene (61.52%), followed by indeno[1,2,3-c,d]pyrene. This value was reduced to 30.56% in the raw food. Benzo[a]anthracene had the second highest contribution in daily consumed raw food.

Acknowledgments

This work was supported by the Ministry of Environmental Protection (No. 200809101) and the Fundamental Research Funds for the Central Universities of China Institute of Industrial Relations (No. 11zy047).

REFERENCES

Brune, H., Deutsch-Wenzel, R, P., Habs, M., Ivankovic, S., Schmähl, D., 1981. Investigation of the tumorigenic response to benzo(a)pyrene in aqueous caffeine solution applied orally to Sprague-Dawley rats. J. Cancer Res. Clin. Oncol. 102, 153–157.

Cejpek, K., Hajšlová, J., Kocourek, V., Tomaniová, M., Cmolik, J., 1998. Changes in PAH levels during production of rapeseed oil. Food Addit. Contam. 15, 563–574.

Ciemniak, A., Chrachol, L., 2008. Polycyclic aromatic hydrocarbons (PAH) in cereal breakfast products. Rocz. Panstw. Zakl. Hig. 59, 301–307.

de Vos, R.H., van Dokkum, W., Schouten, A., de Jong-Berkhout, P., 1990. Polycyclic aromatic hydrocarbons in Dutch total diet samples (1984–1986). Food Chem. Toxicol. 28, 263–268.

Dennis, M.J., Massey, R.C., Mcweeny, D.J., Knowles, M.E., Watson, D., 1983. Analysis of polycyclic aromatic-hydrocarbons in UK total diets. Food Chem. Toxicol. 21, 569–574.

Duedahl-Olesen, L., Christensen, J. H., Højgård, A., Granby, K., Timm-Heinrich, M., 2011. Influence of smoking parameters on the concentration of polycyclic aromatic hydrocarbons (PAHs) in Danish smoked fish. Food Addit. Contam.: Part A 27, 1294–1305.

Ge, K.Y., 1992. The dietary and nutritional status of Chinese population. People's Medical Publishing House, Beijing.

Grung, M., Holth, T.F., Jacobsen, M.R., Hylland, K., 2009. Polycyclic aromatic hydrocarbon (PAH) metabolites in Atlantic cod exposed via water or diet to a synthetic produced water. J. Toxicol. Environ. Health A 72, 254–265.

Hossain, M. A., Hoque, M.Z., 2011. Polycyclic aromatic hydrocarbons in Bangladeshi vegetables and fruits. Food Chem. Toxicol. 49, 244–247.

Howard, J. W., Fazio, T., 1980. Analytical methodology and reported findings of polycyclic aromatic hydrocarbons in foods. J. Assoc. Off. Anal. Chem. 63, 1077–1104.

IARC (International Agency for Research on Cancer), 2004. IARC monographs on the evaluation of carcinogenic risks to humans. Vol. 88. Lyon, France.

Kazerouni, N., Sinha, R., Hsu, C.H., Greenberg, A., Rothman, N., 2001. Analysis of 200 food items for benzo[a]pyrene and estimation of its intake in an epidemiologic study. Food Chem. Toxicol. 39, 423–436.

Kuhn, K., Nowak, B., Behnke, A., Seidel, A., Lampen, A., 2009. Effect-based and chemical analysis of polycyclic aromatic hydrocarbons in smoked meat: a practical food-monitoring approach. Food Addit. Contam. Part A 26, 1104–1112.

Levengood, J. M., Schaeffer, D. J., 2011. Polycyclic aromatic hydrocarbons in fish and crayfish from the Calumet region of southwestern Lake Michigan. Ecotoxicology 20, 1411–1421.

Li, X.R., 2007. Spatial Distribution Pattern of Emission, Dispersion and Exposure of Polycyclic Aromatic Hydrocarbons in Tianjin, China. Peking University, Beijing, China.

Li, X.R., Tao, S., Guo, M., Cao, J., Wang, X.J., Liu, W.X. et al., 2005. Polulation exposure to PAHs in Tianjin Area. Acta Scientiae Circumstantiae 25, 989–993.

Lodovici, M., Dolara, P., Casalini, C., Ciappellano, S., Testolin, G., 1995. Polycyclic aromatic hydrocarbon contamination in the Italian diet. Food Addit. Contam. 12, 703–713.

Lodovici, M., Dolara, P., Casalini, C., Ciappellano, S., Testolin, G., 1995. Polycyclic aromatic hydrocarbon contamination in the Italian diet. Food Addit. Contam. 12, 703–713.

Martí-Cid., Llobet, J.M., Castell, V., Domingo, J.L., 2008. Evolution of the dietary exposure to polycyclic aromatic hydrocarbons in Catalonia, Spain. Food Chem. Toxicol. 46, 3163–3171.

Pan, C.H., Chan, C.C., Wu, K.Y., 2008. Effects on Chinese restaurant workers of exposure to cooking oil fumes: a cautionary note on urinary 8-hydroxy-2'-deoxyguanosine. Cancer Epidemiol. Biom. Prev. 17, 3351–3357.

Perelló, G., Martí-Cid, R., Castell, V., Llobet, J.M., Domingo, J.L., 2009. Concentrations of polybrominated diphenyl ethers, hexachlorobenzene and polycyclic aromatic hydrocarbons in various foodstuffs before and after cooking. Food Chem. Toxicol. 47, 709–715.

Petry, T., Schmid, P., Schlatter, C., 1996. The use of toxic equivalency factors in assessing occupational and environmental health risk associated with exposure to airborne mixtures of polycyclic aromatic hydrocarbons (PAHs). Chemosphere 32, 639–648.

Pfannhauser, W., 1991. Polycyclic aromatic hydrocarbons (PAHs) in food and selected vegetables in Austria. Mitt Geb Lebensmittelunters Hyg. 82, 66–79.

Santos, C., Gomes, A., Roseiro, L.C., 2011. Polycyclic aromatic hydrocarbons incidence in Portuguese traditional smoked meat products. Food Chem. Toxicol. 49, 2343–2347.

Sha, W.Q., 2008. Research of personal PAHs exposure, in: Academic Annual Meeting Collection of Institute of Environmental Science in Liaoning Province. Anshan, China.

Sun, Y., 2007. Analysis of pollution condition of polycyclic aromatic hydrocarbon in food. Chinese J. Health Lab. Tech. 17, 125–127.

Thomson, B., Lake, R., Lill, R., 1996. The contribution of margarine to cancer risk from polycyclic aromatic hydrocarbons in the New Zealand diet. Polycyclic Aromat. Compd. 11, 177–184.

UK FSAo, 2002. PAHs in the UK Diet: 2000 Total Diet Study Samples. FSA.

US EPA, 1991a. Drinking Water Criteria Document for PAH. Prepared by the Office of Health and Environmental Assessment, Environmental Criteria and Assessment Office, Cincinnati, OH for the Office of Water Regulations and Standards, Washington DC.

US EPA, 1991b. Dose-response Analysis of Ingested Benzo[a]pyrene (CAS No. 50-32-8). Human Health Assessment Group, Office of Health and Environmental Assessment, Washington DC.

US EPA, 2001. Integrated Risk Information System (IRIS): Benzo[a]pyrene (CAS No. 50-32-8).

US EPA, 2009. Exposure factor handbook. Office of Research and Development, National Center for Environmental Assessment, Washington DC.

Voutsa, D., Samara, C., 1998. Dietary intake of trace elements and polycyclic aromatic hydrocarbons via vegetables grown in an industrial Greek area. Sci. Total Environ. 218, 203–216.

Wang, L.D., 2005. Chinese national health and nutrition survey report one: Synthesis report. People's Medical Publishing House, Beijing.

Ward, M.H., Sinha, R., Heineman, E.F., Rothman, N., Markin, R., Weisenburger, D. D. et al., 1997. Risk of adenocarcinoma of the stomach and esophagus with meat cooking method and doneness preference. Int. J. Cancer 71, 14–19.

Xia, Z. H., Duan, X.L., Qiu, W.X., Liu, D., Wang, B., Tao, S. et al., 2011. Health risk assessment on dietary exposure to polycyclic aromatic hydrocarbons (PAHs) in Taiyuan, China. Sci. Total Environ. 408, 5331–5337.

Yoon, E., Park, K., Lee, H., Yang, J., Lee, C., 2007. Estimation of excess cancer risk on time-weighted lifetime average daily intake of PAHs from food ingestion. Human Ecol. Risk Assessm. 13, 669–680.

Zhai, F.Y., Yang, X.G., 2006. Chinese national health and nutrition survey report two: Diet and nutrient intake status. People's Medical Publishing House, Beijing.

Zhang, L. L., Lü, J.G., Wu, G.P., Wei, F.S., 2010. Characteristic of daily total exposure to polycyclic aromatic hydrocarbons of people living in coal-fired area in Qujing, Yun'nan Province. China Environ. Sci. 30, 1413–1418.

JOURNAL OF ENVIRONMENTAL SCIENCES

环境科学学报(英文版)

(http://www.jesc.ac.cn)

Aims and scope

Journal of Environmental Sciences is an international academic journal supervised by Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. The journal publishes original, peer-reviewed innovative research and valuable findings in environmental sciences. The types of articles published are research article, critical review, rapid communications, and special issues.

The scope of the journal embraces the treatment processes for natural groundwater, municipal, agricultural and industrial water and wastewaters; physical and chemical methods for limitation of pollutants emission into the atmospheric environment; chemical and biological and phytoremediation of contaminated soil; fate and transport of pollutants in environments; toxicological effects of terrorist chemical release on the natural environment and human health; development of environmental catalysts and materials.

For subscription to electronic edition

Elsevier is responsible for subscription of the journal. Please subscribe to the journal via http://www.elsevier.com/locate/jes.

For subscription to print edition

China: Please contact the customer service, Science Press, 16 Donghuangchenggen North Street, Beijing 100717, China. Tel: +86-10-64017032; E-mail: journal@mail.sciencep.com, or the local post office throughout China (domestic postcode: 2-580).

Outside China: Please order the journal from the Elsevier Customer Service Department at the Regional Sales Office nearest you.

Submission declaration

Submission of an article implies that the work described has not been published previously (except in the form of an abstract or as part of a published lecture or academic thesis), that it is not under consideration for publication elsewhere. The submission should be approved by all authors and tacitly or explicitly by the responsible authorities where the work was carried out. If the manuscript accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

Submission declaration

Submission of the work described has not been published previously (except in the form of an abstract or as part of a published lecture or academic thesis), that it is not under consideration for publication elsewhere. The publication should be approved by all authors and tacitly or explicitly by the responsible authorities where the work was carried out. If the manuscript accepted, it will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

Editorial

Authors should submit manuscript online at http://www.jesc.ac.cn. In case of queries, please contact editorial office, Tel: +86-10-62920553, E-mail: jesc@263.net, jesc@rcees.ac.cn. Instruction to authors is available at http://www.jesc.ac.cn.

Journal of Environmental Sciences (Established in 1989)

Vol. 26 No. 2 2014

Supervised by	Chinese Academy of Sciences	**Published by**	Science Press, Beijing, China
Sponsored by	Research Center for Eco-Environmental		Elsevier Limited, The Netherlands
	Sciences, Chinese Academy of Sciences	**Distributed by**	
Edited by	Editorial Office of Journal of	Domestic	Science Press, 16 Donghuangchenggen
	Environmental Sciences		North Street, Beijing 100717, China
	P. O. Box 2871, Beijing 100085, China		Local Post Offices through China
	Tel: 86-10-62920553; http://www.jesc.ac.cn	Foreign	Elsevier Limited
	E-mail: jesc@263.net, jesc@rcees.ac.cn		http://www.elsevier.com/locate/jes
Editor-in-chief	Hongxiao Tang	**Printed by**	Beijing Beilin Printing House, 100083, China

CN 11-2629/X **Domestic postcode: 2-580** **Domestic price per issue RMB ¥ 110.00**

ISSN 1001-0742