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Distribution and Sources of Polycyclic Aromatic Hydrocarbons in Soils from Shengli Oil Field, China

Shaoping KUANG[1,3], Yaqing SU[2], Jinghuan ZHANG[1], Zhen SONG[1], Huihui WANG[1,3] and Wenjuan YU[2]*

[1]College of Environment and Safety Engineering and [2]College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao, P.R. China;
[3]Shandong Provincial Key Laboratory of Eco-Environmental Science for Yellow River Delta, Binzhou University, Binzhou, P.R. China

**Corresponding author: kuang1009@126.com*

Abstract

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The concentrations of 16 polycyclic aromatic hydrocarbons (PAHs), which are on the United States Environmental Protection Agency (US EPA) priority pollutant list, were studied in ten different soil samples from Shengli Oil Field, China. The total PAHs concentrations in the sampled soils attained 1214.9–2965.1 ng/g (2159.6 ng/g on average). The highest total PAHs concentration was in the soil with a huge content of oil sludge, while the lowest was in fine soil environment areas. The soil contamination with PAHs in the study areas was classified as severe. The major pollutants were naphthalene, phenathrene, fluorine (Flu), pyrene (Pyr), while the detected concentration of benzo(a)pyrene (BaP), benzo(b)fluoranthene was relatively low. Among the 16 kinds of PAHs, the concentration increased in the order: 6 rings < 5 rings < 4 rings < 2 rings < 3 rings. The ratios of Flu/(Flu + Pyr) and indeno benzene(1,2,3-c,d)pyrene (IP)/(IP + BaP) were 0.46–0.48, and 0.36–0.64, respectively. Our results suggest that the main sources of PAHs were petroleum extraction and petroleum combustion. In addition, a small amount of PAHs originated from combustion of grass, woods, and coal.

Keywords: content; oil sludge; PAHs; petroleum extraction; petroleum combustion

The United States Environmental Protection Agency (US EPA) categorized 16 kinds of polycyclic aromatic hydrocarbons (PAHs) in the priority list due to their unfavourable impact on environment and human health, especially the properties of mutagenicity and carcinogenicity (Keiluweit *et al.* 2012). Moreover, PAHs are known to suppress the immune system and are suspected endocrine disruptors. In addition, PAHs could be trapped in soil minerals, while they either leach into the groundwater or escape into the atmosphere, posing a severe risk to flora, fauna, and offsite receptors (Cheng *et al.* 2012). The exploitation of oil field was the main source of oil contaminated soil, which made soil become the most common gathering place for petroleum pollution (Lehrre 1977). Crude oil extraction has become the main pollutant source of polycyclic aromatic hydrocarbons (PAHs) in soil and groundwater. The emission and source of PAHs in the world's industrial countries have been alarming owing to the combustion of petroleum products (Shen *et al.* 2013). PAHs are also released into the environment as a result of natural activities such as terrestrial vegetation synthesis, microbial synthesis, and volcanic activity. Meanwhile, the adverse impact on human body was much worse from the PAHs presence in soil than in air and water (Menzie *et al.* 1992). In addition, low molecular weight PAHs mainly originated from petroleum pollution.

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During past decades, the distribution and sources of PAHs were the central issue of numerous investigations (Barakat *et al.* 2011; Huang *et al.* 2012). The migration route of PAHs in the soil is: pollution source – topsoil – plow bottom layers of the soil – the soil vadose zone under the ground, and underground aquifers. Studies showed that the concentration of PAHs in Greek agricultural land soil was 38–2244 μg/kg, 707 μg/kg on average (Wei *et al.* 2010). The total PAHs concentrations were 23.3–2834 μg/kg in South Korea's rural soil, 38.3–1057 μg/kg in paddy soil, 233–770 μg/kg in Estonian rural soil, and 2200–12 390 μg/kg in urban soil, respectively (Shi *et al.* 2010).

Researchers found that the ratio of low molecular weight PAHs (2–3 rings) to total PAHs in surface soil was small, but increased rapidly with soil depth. In addition, the correlations between the distribution of PAHs in surface and subsurface soils and their own physical and chemical properties were high (Li *et al.* 2011). Several studies demonstrated that total organic carbon (TOC) was the key property influencing PAHs concentration in sediments (Li *et al.* 2012).

Shengli Oil Field is located in the Yellow River delta of Shandong province and it is the second largest oil production base in China. The amount of oil resources is estimated at 145×10^8 t and the exploration area is 19.4×10^4 km^2. The Yellow River delta was established as an efficient ecological economic zone due to its unique geographical location and natural environment. Furthermore, its development and construction attracted a wide attention and support. The oil exploitation areas Gudong and Gudao are located by the Yellow River delta, and their environmental pollution is prevailingly due to the oilfield development, especially the contamination with PAHs. The aims of this study were to detect the content of PAHs in soil and analyze the pollution condition and pollution resources of PAHs in oil mining areas. Therefore, the oil exploitation areas of Gudao and Gudong were selected to analyze the distribution and concentration of polycyclic aromatic hydrocarbons hoping that the results might provide the theoretical basis and be assessed especially by the oil mining companies management and governmental bodies.

MATERIAL AND METHODS

Chemicals and reagents. The chemicals listed below were used in the present research: the mixed standard solution of PAHs (≥ 99%, J&K Chemical Technology Co., Ltd., USA): naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno benzene(1,2,3-c,d) pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene. Phenanthrene and pyrene with deuterium (National Institute of Standards and Technology, Gaithersburg), hexane, methanol, dichloromethane, acetone (Tianjin Kermel Chemical Reagent Co., Ltd., China).

Sample collection and preparation. The soil samples were collected by mixing a plurality of sampling points. Each soil sample consisted of soil from three subsamples selected around each sampling point. The surface soils of the upper 0–25 cm were collected for each of the three subsamples. Then plant debris and stones were removed from the three collected soil subsamples, soil was evenly mixed, excess soil was removed, and 1 kg of soil remained as the sampling point of representative soil samples. Then the collection of samples was put into the disposable folder chain bags, provided with a marker on the number, and inside each disposable folder chain bag a paper with sampling time, location, latitude, and longitude, and the local weather conditions when collecting samples was inserted. After transfer to the laboratory, the samples were refrigerated. The geographic information and properties of the soil samples are listed in Table 1. A simple map of sampling locations and their distribution in the oil field are listed in Figure 1.

The total organic carbon (TOC) content of soils measured on a TOC analyzer (Elementar Vario EL, Elementar, Germany) varied from 0.41 to 1.52%. The soil pH measured at 0.01 M $CaCl_2$ solution ratio of 1:1 (w/v) was nearly 8. The soluble salt percentages of all soil samples varied from 0.064 to 1.692%.

Sample extraction and clean-up. Extraction of PAHs from soil: 20 g soil was added into a 250 ml conical flask with the extraction solvent of 50 ml dichloromethane/acetone (1:1, v/v). The mixture was first treated in ultrasonic cleaner (KQ5200, Kunshan Ultrasonic Instrument Company, Kunshan, China) for 1 h at 25°C. The extraction liquid was removed and replaced with another 50 ml of extraction liquid. The mixture was then retreated for 1 h. The two extracting solutions were put together, and centrifuged at 4000 rpm for 30 min to separate soil and aqueous phases. The supernatant was condensed to 2 ml in a rotary evaporator at 30°C (RV10, Yamato, Japan)

Table 1. Geographic information and properties of analyzed soil samples

No.	North latitude	East longitude	pH	Soluble salt	TOC	Cu	Cr	Zn
				(%)		(mg/kg)		
1	37°50'47''	118°45'15''	8.26	1.692	0.41	45.5	73.5	83.0
2	37°52'43''	118°44'41''	8.09	0.548	1.13	43.9	62.7	84.6
3	37°50'14''	118°45'33''	8.21	0.064	0.97	246.0	65.7	73.6
4	37°49'56''	118°46'02''	8.23	0.288	1.09	122.0	74.7	74.0
5	37°52'57''	118°52'38''	8.56	0.849	0.27	113.0	86.3	84.4
6	37°52'43''	119°05'31''	8.17	0.820	0.36	22.4	88.0	86.7
7	37°55'20''	119°03'24''	8.52	0.968	0.19	131.0	77.4	89.4
8	37°53'07''	119°02'23''	8.61	0.808	1.44	183.0	70.4	76.5
9	37°52'26''	118°52'48''	8.26	0.664	1.52	10.0	86.4	111.0
10	37°50'53''	118°45'02''	8.05	0.452	1.50	0.0	64.5	126.0

TOC – total organic carbon

under the pressure of 450 mbar and rotation speed of 45 rpm. A C18 solid-phase extraction column (Agilent Technologies Co., Ltd., China) was used to perform the process of purification and dehydration. The C18 solid-phase extraction column was first activated with 10 ml methanol, the suitable flow was controlled while adding the extracting liquid, then the top of column was exposed to air and 10 ml dichloromethane/methanol (1:1) as the eluent with the flow rate of 10 ml/min to leach impurity. Dewatering was done using a vacuum pump and 10 ml dichloromethane/hexane (2:3) was used to elute PAHs. Before transferring the concentration liquid to a pear shaped bottle under a pressure of 450 mbar and temperature of 30°C, the bottle was steamed and dried and its walls were thoroughly cleaned with the grade of chromatographic n-hexane. 2 ml deuterated phenanthrene and pyrene were added when the liquid was transferred to the sampling bottle.

Analysis procedure. A gass chromatograph-mass spectrometer Custom GC-MS QP2010 (Shimadzu, Japan) was used to analyze and determine the products. Inlet temperature: 290°C without shunting, sample size: 1 μl, flow quantity of column: 1.0 ml per min (constant current), column temperature: 80°C (2 min) > 20°C/min to 180°C (5 min) > 10°C/min to 290°C (5 min), scanning mode: full scan mode, quality of scanning range: 45–450 amu, ionization energy: 70 EV, quadrupole: 150°C, ion source temperature: 230°C, interface temperature: 280°C, solvent delay time: 5 min, tuning mode: DFTPP.

Quality assurance and quality control. All solvents were of HPLC grade and all glassware were washed in a mixture solution of potassium bichromate and sulfuric acid, ultrasonic cleaning was used. Meanwhile, all washed glassware were dried at 450°C in muffle furnace. Moreover, all experiments were conducted in triplicate to ensure the reproducibility and precision.

Figure 1. A map of sampling locations and their distribution in the oil field

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Risk assessment. The Nemerow index (Chen *et al.* 2016) is simple to calculate and has a clear physical concept. Therefore it is widely used in assessing soil pollution levels. The Nemerow index (P) was calculated as follows:

$$P = \{[(C_i/L_i)^2_{\text{ave}} + (C_i/L_i)^2_{\text{max}}]/2\}_{1/2} \qquad (1)$$

where:

C_i – concentrations of i polycyclic aromatic hydrocarbons

L_i – corresponding standard of i polycyclic aromatic hydrocarbons

According to the Nemerow index, the soil can be classified as clean ($P \leq 0.7$), less clean ($0.7 < P \leq 1.0$), slightly polluted ($1.0 < P \leq 2.0$), medium polluted ($2.0 < P \leq 3.0$), and heavily polluted ($P > 3.0$).

RESULTS AND DISCUSSION

PAHs concentrations in different soils. The concentrations of PAHs of US EPA priority detected in the soils of Gudong and Gudao are presented in Figure 2. The total concentrations of PAHs in all soil samples varied from 1214.9 to 2965.1 ng/g, 2159.6 ng/g on average. The total PAHs concentration was the highest in No. 8 soil and the lowest in No. 5 soil. According to the standard of PAHs concentration in European agricultural soil, soils can be divided into four levels: pollution-free soil (< 200 ng/g), slightly polluted soil (200–600 ng/g), medium polluted soil (600–1000 ng/g), seriously polluted soil (> 1000 ng/g) (Duan *et al.* 2015). In our study, in ten soils the total PAHs concentrations were higher than 1000 ng/g, proving that all soils were seriously polluted by PAHs. The PAHs concentrations at sampling points Nos. 1, 3, 4, 7, 8, and 10 were more than 2000 ng/g, exceeding the heavy pollution limits (1000 ng/g). These challenging results should be taken into account especially by the petroleum companies and government. In addition, as shown in Table 2, all the Nemerow indexes calculated for the ten soil samples exceeded the value of 10, which greatly exceeds the limit of heavy pollution, which means the concentrations of PAHs in that region might be harmful to plants and human.

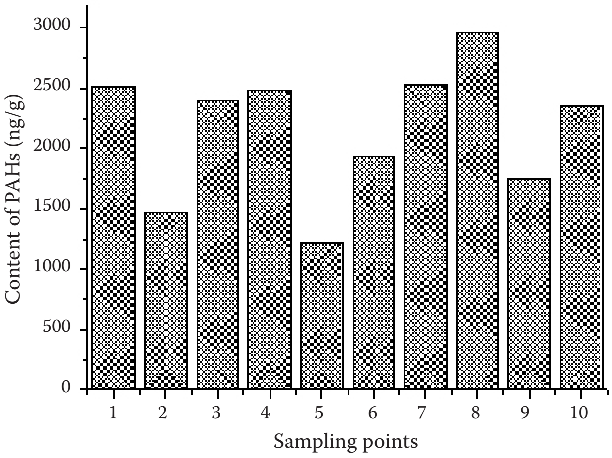

Figure 2. Total polycyclic aromatic hydrocarbons (PAHs) content in different soil samples

The concentrations of all 16 kinds of PAHs in the soils of the study area are presented in Table 3. The concentration of naphthalene was the highest among all the sampling points, varying from 364.7 to 724.3 ng/g, 594.4 ng/g on average, suggesting naphthalene was the main contaminant in each sample, while phenanthrene had the second highest content, ranging from 213.7 to 801.6 ng/g, 436.1 ng/g on average. On the contrary, the concentrations of benzo(a) pyrene and indeno and (1,2,3-c,d)pyrene were the lowest ranging from 10.2 to 26.5 ng/g and 11.7 to 32.9 ng/g, respectively. A previous study showed that the mean concentration of dibenz(a,h)anthracene was the highest in soils around Borholla oilfield – 966 ng/g. And the lowest mean concentration was of benzo(a) pyrene (9.8 ng/g), differing from that in the Shengli Oil Field soils (Jimu *et al.* 2016).

The standard of PAHs in the Canadian Council of Ministers of the Environment's soil quality index (Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health, Soil Quality Index, 2007) was selected as a reference in

Table 2. The Nemerow index (P) of ten analyzed soils

Samples	$(C_i/L_i)^2_{\text{average}}$	$(C_i/L_i)^2_{\text{max}}$	P
1	15.25	203.06	10.45
2	13.62	203.63	10.42
3	15.07	204.20	10.47
4	15.22	204.78	10.49
5	15.11	205.35	10.50
6	13.45	205.92	10.47
7	14.42	206.50	10.51
8	15.54	207.07	10.55
9	16.59	207.65	10.59
10	14.26	208.22	10.55

C_i – concentrations of i polycyclic aromatic hydrocarbons; L_i – corresponding standard of i polycyclic aromatic hydrocarbons

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Table 3. Concentration of 16 kinds of polycyclic aromatic hydrocarbons (PAHs) in ten analyzed soils (in ng/g)

	No. 1	No. 2	No. 3	No. 4	No. 5	No. 6	No. 7	No. 8	No. 9	No. 10
Naphthalene (NaP)	712.5	415.7	678.8	693.5	364.7	561.0	631.0	724.3	497.6	665.1
Acenaphthylene (Any)	109.8	65.1	105.5	106.7	51.1	85.2	97.7	124.8	77.1	101.8
Acenaphthene (Ane)	159.9	92.5	150.1	153.2	75.4	122.1	145.2	169.5	109.8	141.9
Fluroene (Flu)	204.1	119.8	193.2	197.6	96.6	156.3	181.9	213.4	141.5	195.7
Phenanthrene (Phe)	444.9	263.5	429.1	434.8	213.7	344.2	695.6	801.6	310.9	423.1
Anthracene (Ant)	104.4	63.1	101.1	103.3	50.6	82.5	92.6	109.1	74.8	98.0
Fluoranthene (Fla)	110.5	65.0	105.8	107.9	52.3	85.0	95.4	113.6	76.4	102.3
Pyrene (Pyr)	197.7	111.1	188.3	193.3	93.5	146.1	169.0	199.9	135.6	182.6
Benzo(a)anthracene (BaAn)	134.5	76.2	126.5	129.6	62.8	99.7	114.3	134.6	89.9	126.7
Chrysene (Chy)	128.9	77.8	124.7	126.1	61.6	100.9	121.5	145.7	92.5	124.4
Benzo(b)fluoranthene (Bbf)	34.9	21.7	34.1	33.5	16.8	26.6	30.6	36.9	25.1	34.1
Benzo(k)fluoranthene (Bkf)	40.2	23.8	28.0	38.1	18.5	29.4	34.8	42.3	27.6	35.5
Benzo(a)pyrene (BaP)	23.5	14.7	22.6	23.1	10.2	17.2	21.2	26.5	17.2	24.0
Indeno(1,2,3-c,d)pyrene (I1P)	28.7	17.9	27.6	28.2	11.7	21.1	26.9	32.9	20.7	28.6
Benzo(a,h)anthracene (Daa)	33.6	20.8	32.4	32.9	15.0	24.9	30.7	38.8	24.0	32.9
Benzo(g,h,i)perylene (BgP)	47.7	28.3	45.1	46.2	20.4	34.4	42.3	51.2	32.6	44.8
Σ_{16}PAHs	2515.8	1477	2392.9	248	1214.9	1936.6	2530.7	2965.1	1753.3	2361.5

this study since China has no related standards on PAHs in soil. As shown in Figure 3, compared to the Canadian standard (Table 4), the concentration of naphthalene was about 12 times higher, which was the most from all the PAHs. The concentrations of acenaphthene, anthracene, and fluorene exceeded 1–4 times, while the concentrations of anthracene, benzo(a)anthracene, and chrysene were close to the standard's upper limit. Meanwhile, the concentrations of fluoranthene, pyrene and benzo(b)fluoranthene, benzo(a)fluoranthene and benzo(a)pyrene, indeno and (1,2,3-c,d) pyrene, benzo (g,h,i) perylene were below the standard's upper limit.

To further investigate the PAHs distribution, the composition profiles of PAHs with a different number of benzene rings are shown in Figure 4. The 3 rings PAHs accounting for 40.12–47.93% (42.12% on average) were the dominant PAH compound in the soil samples. The 2 rings PAHs ranged from 24.43 to 30.02% (27.81% on average), while the 4 rings PAHs from 19.77 to 22.79% (22.01% on average). The 5 rings PAHs occupied from 11.31 to 13.11% (12.64% on average). In addition, the 6 rings PAHs ranged from 2.64 to 3.13%, the values being the lowest of all the sampling points. Similarly the detected percentages of 2-, 3-, 4-, 5-, and 6-rings PAHs to the total PAHs were 24.43, 35.88, 24.18, 10.7, and 4.8% respectively in 2012, and 19.1, 44.97, 23.53, 8.77, and 3.64% in 2013 in soils from the water-level-fluctuation zone of the Three Gorges Reservoir, China (Hu *et al.* 2017). Hence, the composition of total PAHs in soil was 6 rings < 5 rings < 4 rings < 2 rings < 3 rings at both sites. Concerning the sum of low molecular weight (LMW, 2–3 benzenoid rings), PAHs ranged between 68.84 and 72.87%, 69.93% on average, while as to the high molecular weight (HMW, 4–6 benzenoid rings), PAHs sum was < 39% of the total PAHs at all sampling sites. This suggests that the composition pattern of PAHs in the Shengli Oil field was dominated by LMW. The PAHs composition pattern in this study is similar to that in most studies (Zhao *et al.* 2014; Li *et al.* 2015; Hong *et al.* 2016; Zhang *et al.* 2016). In contrast, the mean percentages of LMW

Table 4. The Canadian Soil Quality standard

No.	Name	Ring value	Standard (mg/kg)
1	naphthalene (Nap)	2	0.05
2	acenaphthene (Ace)	3	0.05
3	acenaphthylene (Any)	3	0.08
4	fluroene (Flu)	3	0.05
5	phenanthrene (Phe)	3	0.19
6	anthracene (Ant)	3	0.05
7	fluoranthene (Fla)	4	0.24
8	pyrene (Pyr)	4	0.19
9	benzo(a)anthracene (BaAn)	4	0.1
10	chrysene (Chy)	4	0.1
11	benzo(b)fluoranthene (Bbf)	5	0.3
12	benzo(k)fluoranthene (Bkf)	5	0.05
13	benzo(a)pyrene (BaP)	5	0.1
14	indeno(1,2,3-c,d)pyrene (I1P)	6	0.11
15	benzo(a,h)anthracene (Daa)	5	0.15
16	benzo(g,h,i)perylene (BgP)	6	0.2

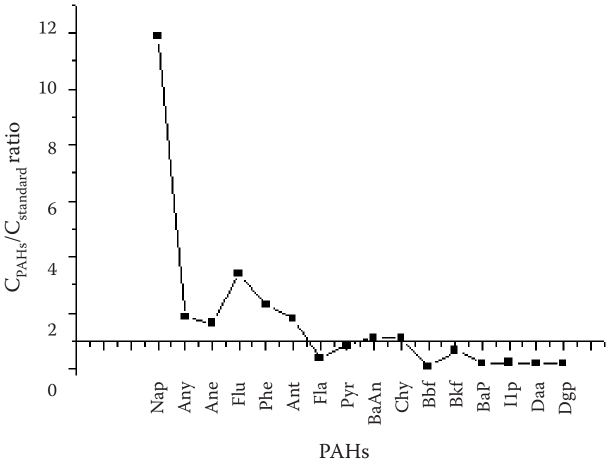

Figure 3. The $C_{PAHs}/C_{standard}$ ratio in soil samples

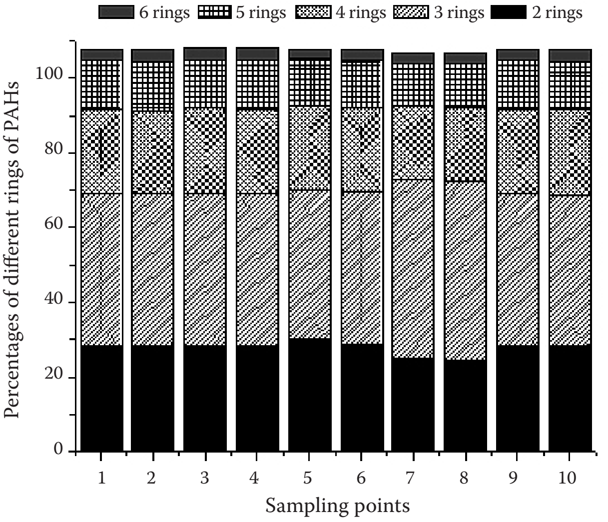

Figure 4. Percentages of different rings of polycyclic aromatic hydrocarbons (PAHs)

Table 5. The ratio of Flu/(Flu+Pyr) and IP/(IP+BgP) in ten analyzed soils (explain the abbreviations)

No.	1	2	3	4	5	6	7	8	9	10
IP/(IP+BgP)	0.38	0.63	0.61	0.38	0.36	0.38	0.39	0.64	0.39	0.39
Flu/(Flu+Pyr)	0.46	0.49	0.46	0.46	0.46	0.46	0.48	0.46	0.47	0.46

Flu – fluorine; Pyr – pyrene; IP – indeno benzene(1,2,3-c,d)pyrene; BgP – benzo(g,h,i)perylene

and HMW PAHs in Brisbane River sediments were less than 25% and over 70% (Duodu *et al.* 2016).

Sources of PAHs in soils. PAHs occurred in high amounts throughout the environment, being released mainly during an incomplete combustion of fossil fuels and organic matter (Timoney & Lee 2011; Zhang *et al.* 2012; Katsoyiannis & Breivik 2014). In this study, the ratio of fluorine (Flu)/(Flu + pyrene (Pyr)) was used to determine the sources of PAHs pollution in soil. The Flu/(Flu + Pyr) ratio < 0.4 was attributed to petrogenic source, ratio > 0.5 to wood and coal combustion, 0.4–0.5 to petroleum combustion (Dvorska *et al.* 2011). The ratios of indeno benzene(1,2,3-c,d) pyrene (IP)/(IP + benzo(a)pyrene (BgP)) < 0.2 indicated petrolenic and petroleum sources of pollution, 0.2 < IP/(IP + BgP) < 0.5 indicated petroleum combustion (including liquid fossil fuels, vehicle, and crude oil combustion), and IP/(IP + BgP) > 0.5 indicated that the source of PAHs are biomass and coal combustion (Suma *et al.* 2016). As shown in Table 5, in this study, the ratio of Flu/(Flu+Pyr) ranged from 0.46 to 0.48, 0.47 on average, and the ratio of IP/(IP + BgP) ranged from 0.36 to 0.64, 0.46 on average. These results indicated that the main source of PAHs in soils of the region was petroleum pollution and petroleum combustion. Due to their various sources, the PAHs differed in structure and composition and remained relatively stable in the process of migration and deposition. Therefore, the pollution sources could be distinguished by component characteristics of polycyclic aromatic hydrocarbons.

Concerning the environment information of the soil samples, sampling points No. 2 and No. 5 showed a low concentration of PAHs in soil, because point No. 2 was surrounded by farmland, No. 5 was near a reservoir and wetlands. The concentrations of PAHs at the sampling points Nos. 1, 3, 4, 7, 8, and 10 were higher than at the rest of them. All the spots above were around oil drillings. And the possible resources of the soil environment pollution were oil well, sludge, and a sewage treatment plant.

Relation of PAHs concentration and TOC. TOC is a key property influencing PAHs concentration, and sedimentary TOC is a crucial factor determining the sorption, sequestration, and fate of PAHs (Mai *et al.* 2002). In this study, the correlation between the concentrations of PAHs and TOC contents were calculated through the Pearson's correlation analysis (Gu *et al.* 2016). Figure 5 shows the correlation between the total PAHs concentrations and the TOC percentages, indicating that there was an insignificant correlation between the concentrations of PAHs and the TOC content. Poor correlations between PAHs and TOC were reported in previous studies, too (Gu *et al.* 2013).

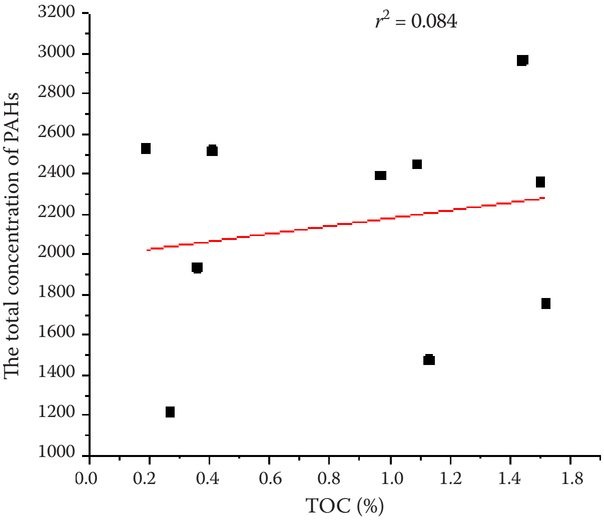

Figure 5. Correlation between total polycyclic aromatic hydrocarbons (PAHs) and total organic carbon (TOC)

CONCLUSION

PAHs concentrations were relatively high in the study region, with an average of 2159.58 ng/g. The main source of PAHs in the study region is petroleum and all the pollution by PAHs in the soil sampling sites could be categorized as heavy. Moreover, the dominant PAHs detected in the soil of all the sampling areas were found to be naphthalene, phenathrene, fluorene, pyrene, and benzo(a)pyrene. The concentrations of PAHs in drilling, sludge or sewage treatment plant were higher than in other places. To conclude, the results of the present investigation provide useful information to assess the PAHs concentration levels in the Shengli Oil Field.

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