



Occurrence, sources and health risk of polycyclic aromatic hydrocarbons in soils around oil wells in the border regions between oil fields and suburbs

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ARTICLE INFO

Keywords:

PAHs
Oil extraction history
Source apportionment
Carcinogenic and non-carcinogenic risks

ABSTRACT

The Yellow River Delta (YRD) is a typical region where oil fields generally overlap cities and towns, leading to complex soil contamination from both the oil fields and human activities. To clarify the distribution, speciation, potential sources and health risk of polycyclic aromatic hydrocarbons (PAHs) in soils of border regions between oil fields and suburbs of the YRD, 138 soil samples (0–20 cm) were collected among 12 sampling sites located around oil wells with different extraction histories. The 16 priority control PAHs (16PAHs), as selected by the United States Environmental Protection Agency (USEPA), were extracted via an accelerated solvent extraction and detected by GC-MS. The results showed that soils of the study area were generally polluted by the 16PAHs. Among these pollutions, chrysene and phenanthrene were the dominant components, and 4-ring PAHs were the most abundant. A typical temporal distribution pattern of the 16PAHs was revealed in soils from different sampling sites around oil wells with different exploitation histories. The concentrations of total 16PAHs and high-ring PAHs (HPAHs) both increased with the extraction time of the nearby oil wells. Individual PAH ratios and PCA method revealed that the 16PAHs in soil with newly developed oil wells were mainly from petroleum pollutants, whereas PAHs in soils around oil wells with a long exploitation history were probably from petroleum contamination; combustion of petroleum, fuel, and biomass; and degradation and migration of PAHs from petroleum. Monte Carlo simulation was used to evaluate the health risks of the 7 carcinogenic PAHs and 9 non-carcinogenic PAHs in the study area. The results indicated that ingestion and dermal contact were the predominant pathways of exposure to PAH residues in soils. Both the carcinogenic and non-carcinogenic burden of the 16PAHs in soils of the oil field increased significantly with exploitation time of nearby oil wells.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are typical persistent organic pollutants (POPs) that have borne decades of attention by environmental chemists and toxicologists. Among the many existing PAHs, the United States Environmental Protection Agency (USEPA) identified 16 as priority pollutants (16PAHs). Owing to their toxic, mutagenic, carcinogenic and degradation-resistance characteristics, it is important that the concentration, distribution, potential sources and health risk of the 16PAHs in soils be clarified (Ding et al., 2012; Jiang et al., 2009; Oyo-Ita and Oyo-Ita, 2013; Wang et al., 2006; Wang et al., 2011). Generally, high PAH concentrations in soil environments are from petroleum contamination; incomplete combustion and pyrolysis of organic materials of traffic, industry, and agriculture; long-term irrigation with wastewater; and use of sewage sludge and fertilizer in agriculture (Agarwal et al., 2009; Fang et al., 2014; Hezhong et al., 2015).

The Shengli Oil Field, which is located in the Yellow River Delta

(YRD), is the second biggest oil field in China and has been exploited for almost 60 years since 1961. Decades of crude oil exploitation of this oil field have led to complex contamination by petroleum hydrocarbons and PAHs, which are the main constituents of crude oil (Fu et al., 2014; Wang et al., 2011; Xie et al., 2012; Yuan et al., 2014, 2015). Cities, towns, and villages in this region evolved simultaneously with the development of the oil field, which has led to large areas, inhabited by millions of people (Buttafuoco et al., 2017; Guagliardi et al., 2012, 2016, 2018), in the border regions between the oil field and nearby suburbs. The health risks of PAHs for the people exposed to PAHs in the border regions cannot be ignored (Li and Boufadel, 2010; Ribeiro et al., 2013; Zhu et al., 2015).

However, few researches exist on the relationships between the PAHs in soils and the exploitation history of oil wells nearby. The distribution, speciation and potential sources of PAHs in soils of the border region between oil fields and suburbs, which could be quite specific and influenced during the extraction of crude oil, remain ambiguous. Furthermore, accurate and scientific health risk assessment should also

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be carried out, in order to clarify the actual human health risk of exposure to soil PAHs in border regions of oil fields. In this study, surface soil samples at 12 sampling sites ($n = 138$) around oil wells with different exploitation histories were collected and analyzed to clarify the temporal distribution, speciation and potential sources of the 16PAHs in the border region between an oil field of the YRD and suburbs nearby. The cancer risk (CR) and hazard quotient of from direct exposure to PAHs in soils around oil wells with different exploitation history was calculated using the Monte Carlo method to estimate the excess cancer risk due to intake of the targeted chemicals in the study area.

2. Materials and methods

2.1. Soil sampling and preparation

The YRD is the primary region of the Shengli Oil Field of China. This oil field straddles about eight cities of Shandong Province in China, covers an area of about 61,000 km², and has been scattered with more than 20 thousand oil wells since 1995. The study area is located in the East Asian monsoon region, with an annual temperature of about 12.1 °C and precipitation of 580 mm. The soil types are dominated by saline flava-aquic soil and coastal alluvial soil, according to Chinese soil taxonomy (CST).

The city of Dongying and the town of Gudao are located in the middle of both the YRD and the Shengli Oil Field. These entities evolved simultaneously with the development of the oil field, leading to the situation in that thousands of oil wells are scattered in border regions between the oil field and nearby suburbs.

Surface soil samples (0–20 cm) were collected in May 2015 in typical border regions between the Shengli Oil Field and the suburbs of the city of Dongying and the town of Gudao in Shandong Province, China (shown in Fig. 1). The sample collection area targeted belongs to the oil production regions of Gudao, Xianhe, and Dongxin of the Shengli Oil Field.

In each of the three targeted oil production regions, four sampling sites were selected around oil wells with different initial exploitation times, once each in the 2010s, 2000s, 1990s, and 1980s (Table 1). About 10 soil samples were collected at each sampling site (the exact numbers of samples are shown in Table 1). The number collected depended on the conditions of the sampling sites. As a result, 138 samples among 12 sampling sites were collected in this study. On average, five subsamples of equal amounts from the same sampling point were homogeneously mixed. Approximately 1 kg of the mixed sample was used to represent the sample point. The control point (CP) was also shown in the Fig. 1.

All soil samples were collected within close proximity to oil wells, but away from asphalt, tar blocks, and obvious oil spillages into the soil. Each soil sample was divided into two portions: an aluminum box was filled with one and the second was stored at about 4 °C in a glass bottle. All soil samples were freeze-dried and finely ground to pass through an 80-mesh sieve after impurities were discarded.

2.2. Analytical methods

Using accelerated solvent extraction (ASE-300, Dionex, USA), PAHs in each soil sample (about 20 g) were pressurized-liquid extracted with an extraction solvent of dichloromethane/*n*-hexane (4:1, v/v). The pressure of the extraction cell was raised to 10.3 MPa (1500 psi), and the temperature was increased to 100 °C. The heating process and the static extraction process were each held for 5 min, respectively. Then, the extraction cell was flushed with the extraction solvent (60% of the cell volume) and blown with air for 60 s. Finally, the PAHs were obtained after the extraction was solvent-replaced with dichloromethane and concentrated to 10 mL.

The PAHs in the soil sample extractions were all analyzed via a gas

chromatograph/mass spectrometer [7890A GC/5975C MS, Agilent, USA] equipped with an HP-5MS capillary column (30 m length, 0.25 mm inner diameter, 0.25 μm film thickness; Agilent, USA). The carrier gas was helium (high purity, 99.99%) at a flow rate of 1 mL min⁻¹. The temperature of the injection port was 280 °C. The temperature of the oven was initially set at 60 °C and held for 1 min, and then increased at a rate of 20 °C/min to 150 °C, where it was held for 8 min. After that, the oven temperature was raised at a rate of 5 °C/min to 270 °C, where it was held for 3.36 min, and finally, at a rate of 10 °C/min, it reached 290 °C, where it was held for 10 min. The injection volume was 1 μL, with unsplit stream sampling. The mass spectrometric detector (MSD) was operated under the electron impact mode at 70 eV, with the temperature of the ion source at 230 °C and the quadrupole rod at 150 °C. To identify the PAHs, the MSD was operated with a scan mode ranging from 60 to 640 amu.

The concentrations of the PAHs were represented by the quantification of the 16PAHs, based on the peak area external reference method with a mixed PAH standard (J&K Co., USA): naphthalene (Nap; 2-ring), acenaphthylene (Ace; 3-ring), acenaphthene (Acy; 3-ring), fluorene (Flu; 3-ring), phenanthrene (Phe; 3-ring), anthracene (Ant; 3-ring), fluoranthene (Fle; 4-ring), pyrene (Pyr; 4-ring), benzo(a)anthracene (BaA; 4-ring), chrysene (Chr; 4-ring), benzo(b)fluoranthene (BbF; 5-ring), benzo(k)fluoranthene (BkF; 5-ring), benzo(a)pyrene (BaP; 5-ring), dibenz(a,h)anthracene (DBA; 5-ring), indeno(1,2,3-c,d)pyrene (InP; 6-ring), and benzo(g,h,i)perylene (BgP; 6-ring). ΣPAHs indicates the sum concentration of the 16 PAHs within a sample. The peak area was plotted against the standard reference every 5 days to construct the calibration graphs. The R² was more than 0.997, indicating a linear relationship. The method detection limits of PAHs ranged from 0.8 ng g⁻¹ (2-ring) to 7.3 ng g⁻¹ (6-ring). The recoveries ranged from 82.8% to 94.2%, with an average of 81%, and the relative standard deviation (RSD) was lower than 8.86%.

The standard sample was prepared by adding the 16PAH standards, which conform to the quality control acceptance criteria of the USEPA (USEPA, 2007). Blank samples were extracted and determined under the same conditions. Because the results of the blanks were all below the detection limits, the determination results of the 16PAHs were presented without blank corrections in this paper.

2.3. Health assessment method

To evaluate the probabilistic risk of excess cancer due to people's direct exposure to PAHs from soils, the cancer risk (CR) and non-carcinogenic risk (hazard quotient, HQ) were both estimated. The exposure was estimated by calculating three intake routes: soil ingestion, dermal contact and inhalation. The daily dose (ADD, mg (kg d)⁻¹) of the pollutant by adult inhabitant via three exposure pathways can be estimated by Eqs. (1)–(3) below (Chen and Liao, 2006; Li and Li, 2017; Hu et al., 2014). The CR and HQ values can be estimated using Eqs. (4) and (5), respectively.

$$\text{ADD}_{\text{ingest}} = \frac{C_s \times \text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (1)$$

$$\text{ADD}_{\text{dermal}} = \frac{C_s \times \text{SA} \times \text{AF} \times \text{ABS} \times \text{EF} \times \text{ED} \times \text{CF}}{\text{BW} \times \text{AT}} \quad (2)$$

$$\text{ADD}_{\text{inhale}} = \frac{C_s \times \text{InhR} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BW} \times \text{AT}} \quad (3)$$

$$\text{CR} = \sum (\text{ADD}_i \times \text{SFi}) \quad (4)$$

$$\text{HQ} = \sum (\text{ADD}_i / \text{RfDi}) \quad (5)$$

where C_s is the concentration of targeted chemicals in the soil (ng g⁻¹); IngR is the soil ingestion rate, 100 mg d⁻¹; EF is the exposure frequency, 350 d year⁻¹; ED is the exposure duration, 30 years; BW is the average body weight, 58.78 kg; AT is the averaging time, 25,550 days

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