Journal of Environmental Management 174 (2016) 55–61

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Consistency between health risks and microbial response mechanism of various petroleum components in a typical wastewater-irrigated farmland

Juan Zhang [a, b, *], Shu-kai Fan [c]

[a] School of Metallurgical and Ecological Engineering, University of Science and Technology Beijing, Beijing, 100083, PR China
[b] Beijing Key Laboratory of Green Recycling and Extraction of Metal, Beijing, 100083, PR China
[c] Environmental Engineering Institute, Beijing General Research Institute of Mining and Metallurgy, Beijing, 100160, PR China

ARTICLE INFO

Article history:
Received 23 November 2015
Received in revised form
24 February 2016
Accepted 13 March 2016
Available online 21 March 2016

Keywords:
Bacterial diversity
Membrane fluidity
Bacterial phospholipid fatty acids
Health risk models
Multivariate statistics

ABSTRACT

Various petroleum components possess distinctive migration and toxicity characteristics. Evaluation of contamination levels on the basis of total concentrations of petroleum hydrocarbons in soil and groundwater is limited. Hunpu, a typical wastewater-irrigated area, is located at the southwest of Shenyang City, Liaoning Province, China. In this study, various fractions, exposure pathways, and soil microbial communities were taken into account to make petroleum contamination evaluation more effective and precise in the region. The concentrations and hazard quotients of aliphatic fractions, as the bulk of an oil, verified that the groundwater must not be drunk directly. The total concentrations of aliphatic hydrocarbons (TAHs) for C_{10-34} were 68.90–199.87 μg g^{-1} in soil in Hunpu, which required cleanup according to Oklahoma criteria. However, both health and ecological risks indicated that petroleum contamination in surface soil was not serious. Microbes may use aliphatic fractions as carbon and energy source for their growth, which was indicated by positive correlation between them. TAHsC_{12-16} posed highest human health risks and had the most significant effect on the soil microbial composition, although its concentration was low in both the groundwater and the soil. Straight-, branched-chain saturated, and cyclopropyl phospholipid fatty acids had more closely positive correlation with TAHsC_{12-16}, which indicated that regulation of bacterial membrane fluidity to toxic petroleum pollutants. This study can also provide the guidelines for assessment and management of petroleum contamination.

1. Introduction

Aliphatic fractions constitute the bulk of an oil, and can be used to assess total oil concentrations (Douglas et al., 1996; Singh et al., 2012). Aliphatic hydrocarbons pose risks to human health and soil microbes. These compounds are more likely to be sequestered in the epidermis than are aromatic hydrocarbons (Baynes et al., 2000; McDougal et al., 2000). They are responsible for pneumonia by inhalation, folliculitis by skin contact, paraffinoma or oleogranuloma by subcutaneous injection, follicular lipidosis by chronic ingestion, and the accumulation of long-chain in viscera (Chou et al., 2002; Salvayre et al., 1988). The hazard quotients (HQ) of the US Environmental Protection Agency (EPA) are extensively used to characterize the noncarcinogenic health effects of pollutants by comparison of their effects from exposure to a reference dose (RfD) (ATSDR, 1999; Qu et al., 2012). Aliphatic hydrocarbons are divided into TPH-Aliph > C_{10-12}, TPH-Aliph > C_{12-16}, TPH-Aliph > C_{16-21}, and TPH-Aliph > C_{21-34} according to migration and toxicity difference (Connor et al., 2007; TCEQ, 2009).

Furthermore, it is important to combine both biological responses and chemical analysis for integrated environmental assessment because of limitation of chemical monitoring (Blasco and Picó, 2009). The uses of biological endpoints increase, which help to appropriately define acceptable cleanup standards of contaminated sites and establish the ecological soil quality assessment (Płaza et al., 2008). Microbial communities present important functions in soil because of their contribution in nutrient cycling, plant symbioses, decomposition, and other ecosystem

* Corresponding author. School of Metallurgical and Ecological Engineering, University of Science and Technology Beijing, 30 Xueyuan Road, Haidian District, Beijing, 100083, PR China.
E-mail addresses: zhangjuan85@ustb.edu.cn, sikao-xiwang@163.com (J. Zhang), fanshukai@bgrimm.com (S.-k. Fan).

http://dx.doi.org/10.1016/j.jenvman.2016.03.018
0301-4797/

processes. And they are sensitive to petroleum hydrocarbon contamination in the soil. As a culture-independent, well-characterized and convenient technique, phospholipid fatty acid (PLFA) analysis has been demonstrated to be suitable to profile microbial community structure response to petroleum hydrocarbon contamination (Piotrowska-Seget and Mrozik, 2003; Schafer et al., 2009; Wei et al., 2014; Zhang et al., 2012a).

Aliphatic hydrocarbons concentrations have been widely used in environmental evaluation and management of petroleum contamination. There is growing awareness of the limitation of concentrations. The assessment models of potential human health risks and the parameters related to aliphatic hydrocarbons have been proposed by environmental organizations and agents in many countries (ASTM, 2015; Connor et al., 2007; China MEP, 2014). Moreover, the effects of petroleum hydrocarbon contamination on microbial community structure have been focused on. Comprehensive evaluation has not been reported using concentrations, the forecasting health risks, and their effects on microbial community structure in the contaminated soil under complex natural conditions. And consistency of results from these methods has not yet discussed. It can give guidelines to select effective evaluation methods and build appropriate criteria not only for aliphatic hydrocarbons but also for other contaminants. The mechanism of microbial response to petroleum contamination can also be revealed using PLFAs, indicative of membrane structure.

Shenyang is the most important industrial city that serves as the economic and cultural center in Northeastern China. The Xihe River is the major drainage channel for the city's industrial and municipal sewage. The Xihe River, its riverbank groundwater, and the Hunhe River are severely contaminated with petroleum hydrocarbons (Guo et al., 2011). For more than 40 years, the Hunpu region has been irrigated with the wastewater, which resulted in the excessive accumulation of organic pollutants in agricultural soils and groundwater (Song et al., 2005; Zhang et al., 2012b). In addition, the point source pollution and the petroleum hydrocarbons in the air directly entering the soil by atmospheric deposition could enhance aliphatic hydrocarbons concentrations in the Hunpu. Therefore, the region was selected as the studied area.

This study aims to evaluate effectively the health risks of four aliphatic fractions (TPH-Aliph > $C_{10–12}$, TPH-Aliph > $C_{12–16}$, TPH-Aliph > $C_{16–21}$, and TPH-Aliph > $C_{21–34}$) in the soil and groundwater using contaminant migration, diffusion, and exposure risk assessment models. And then it aims to explore the response and possible mechanisms of soil microbial communities to different aliphatic fractions using multivariate statistical analysis based on PLFAs. Finally, consistency and disagreement between various environmental evaluation methods including concentrations, human health risks, and ecological responses will be revealed.

2. Material and methods

2.1. Sampling

In October 2010, soil and groundwater samples were collected at the Hunpu wastewater-irrigated area located at the southwest of Shenyang City, Liaoning Province, China. The Hunpu region has an area of 410,000 ha with a typical temperate monsoon climate. The annual mean temperature is 7.8 °C, and the annual mean precipitation is 734.4 mm. The predominant wind direction all year is toward the south. The soil texture is defined as sandy loam and the soil taxonomy is defined as a vertisol. Irrigation water is delivered by the main canal and the Xihe River canal, both of which draw water from the Hunhe River (Fig. 1). The Xihe River is the major drainage channel for the industrial and municipal sewage of Shenyang. The main canal has some sub-main canals and connects to the Xihe River canal through its fourth sub-main canal.

Considering the effect of wastewater, six soil units (paddy fields: I-1P, I-3P, and I-5P; upland fields: I-1U, I-4U, and I-6U) along the strike of the irrigation canals were selected (Fig. 1). Five 100 m × 100 m blocks far away from the roads (each block included single or multiple fields) were randomly selected at each unit. Each surface soil sample (0–10 cm below ground surface) was homogeneously mixed with five diagonally sampled subsamples in each block. A portion of each sample was filled into a pre-cleaned aluminum box and transported to the laboratory at a temperature of 4 °C. The samples were freeze-dried and ground to pass through 1 mm mesh for petroleum hydrocarbons and PLFA analyses.

Three corresponding wells were randomly selected at each unit. Before sampling, each well was purged. Each sample was filled into two 500 mL brown glass jars with no headspace. The samples were sealed with Teflon film and then transported to the laboratory at a temperature of 4 °C. An aliquot of 10–30 mg mercuric chloride was added to each sample to retard microbial activity (Christian and Karl, 1995). All sample wells were bore wells with a diameter of 7–8 cm. The wells were confined wells, with depths to water of 50–70 m at I-1P and I-1U. The wells were unconfined wells, with depths to water of 20–30 m at I-3P and I-4U. The wells were unconfined wells, with depths to water of approximately 10 m at I-5P and I-6U.

2.2. Aliphatic hydrocarbon analysis

For extraction of petroleum hydrocarbons in soil, a soil sample (approximately 15 g) was soxhlet extracted with 300 mL CH_2Cl_2 for 24 h (USEPA, 1996a). Prior to extraction of petroleum hydrocarbons in water, the pH of the sample was adjusted to 2 using 1:1 (V/V) H_2SO_4. About 30 g of NaCl was added to the extraction system to further avoid emulsification. And then, an aliquot of approximately 500 mL of groundwater was subjected to separatory funnel liquid-liquid extraction with 30 mL CH_2Cl_2 × 4 (USEPA, 1996b). The separatory funnel was then sealed and shaken vigorously for 10 min with periodic venting. The separatory funnel was laid flat and ultrasonic vibrated or heated if emulsification occurred. The resulting extract was then cooled and concentrated to 2 mL using a rotary vacuum evaporator (35 °C). The aliphatic hydrocarbons were obtained through eluting with approximately 20 mL n-hexane after purification with an alumina and silica gel chromatography column and concentrated to 1 mL for qualitative and quantitative analyses (Van De Weghe et al., 2006).

The aliphatic hydrocarbon fraction was analyzed by an Agilent 7890A GC with a HP-5 fused silica capillary column (30 m × 0.25 mm i.d. × 0.25 mm film thickness). A 2 μL volume was injected in the pulsed splitless mode. Oven temperature was initially at 60 °C for 2 min and then increased to 320 °C at 8 °C min^{-1} for 10 min. The flame ionization detector (FID) was at 330 °C. The carrier gas was N_2. *N*-alkanes (nC_{10} to nC_{40}), phytane, and pristane were quantified using the external standard (Accustandard, US) method. Unresolved complex mixture (UCM) was quantified by assuming a response factor of 1.0 based on nonadecane. The total aliphatic hydrocarbons (TAHs) value for > $C_{10–12}$, > $C_{12–16}$, > $C_{16–21}$, and > $C_{21–34}$ was the sum of the n-alkanes and UCM from nC_{10} to nC_{12}, from nC_{12} to nC_{16}, from nC_{16} to nC_{21}, and from nC_{21} to nC_{34}, respectively.

The analytical procedure was rigidly evaluated. Calibration graphs were constructed by plotting the peak area against the reference material concentration every two days. A linear relationship with $r^2 \geq 0.999$ was obtained. The recovery test was performed with a reference material-spiked soil. A control soil sample (approximately 15 g) was spiked with three kinds of reference material solutions with concentrations of 2, 6, and 10 μg ml^{-1},