

Spatial Distribution Patterns and Potential Sources of Heavy Metals in Soils of a Crude Oil-Polluted Region in China*¹

FU Xiao-Wen, WANG Deng-Ge, REN Xiao-Hua and CUI Zhao-Jie*²

School of Environmental Science and Engineering, Shandong University, Ji'nan 250100 (China)

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ABSTRACT

Twenty-two soil samples were collected at the subregional scale (50 km²) around Gudao Town, a typical oil-producing region of the Shengli Oilfield in the Yellow River Delta, China to determine the spatial distribution patterns and potential sources of heavy metals in soils of crude oil-polluted regions. The concentrations of total petroleum hydrocarbons (TPHs) and heavy metals as well as other soil properties were determined and the enrichment factor values were calculated for the heavy metals measured. Principal component analysis (PCA) and cluster analysis (CA) were used to estimate potential sources contributing to the concentrations of heavy metals in the soils. The results revealed that the soils were slightly alkaline (pH = 7.33–8.05) and high in salinity (1.43–41.30 g kg⁻¹), TPHs (0.51–28.40 g kg⁻¹) and organic matter (1.74–31.50 g kg⁻¹). The mean concentrations of the measured heavy metals Cu, Zn, Pb, Cd, Cr, Ni and V were 18.4, 78.2, 20.8, 0.19, 56.6, 26.3 and 62.1 mg kg⁻¹, respectively. Although the concentrations of all the metals measured in this study were not high enough to exceed the national control standards, there was a significant enrichment of Cd in the study area and Zn and Ni were in the category of deficiency to minimal enrichment. The spatial distribution patterns of Cu, Cr, Ni and V were similar and partially affected by oil exploitation and petroleum hydrocarbon spills. Potential sources of Cr, Ni, V and Cu in the soils were both natural sources and petroleum hydrocarbon spills, while Zn, Pb and Cd were probably from anthropogenic sources such as farming activities and traffic.

Key Words: cluster analysis, enrichment factor, farming activities, petroleum hydrocarbon spills, principal component analysis

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INTRODUCTION

Because of their severe toxicity and wide distribution in the soil environment, heavy metals have been regarded as some of the most dangerous soil pollutants (Al-Muzaini and Jacob, 1996; Facchinelli *et al.*, 2001; Gallego *et al.*, 2002). The soil heavy metal pollution is a serious threat because the heavy metals may adversely affect the soil ecosystem and be transferred to groundwater, crops and human beings at the end of the food chain (Roane and Kellogg, 1996; Kelly and Tate, 1998; Mikanova, 2006; Ruan *et al.*, 2008; Chen *et al.*, 2009). The potential sources of heavy metals in soils can be generally divided into two groups: natural and anthropogenic sources (Micó *et al.*, 2006; Zhao *et al.*, 2010; Wu *et al.*, 2011). Clarifying potential sources and distributions of metals after they enter soils is extremely important for environmental management and pollution control (Liu *et al.*, 2007; Zhao *et al.*, 2007). Previous research has reported that crude oil contains heavy metals such as Ni and V (Yen,

1975; Krzyzanowski, 2012), and oil pollution (spills of petroleum hydrocarbons) could significantly affect the distribution of heavy metals in soil (Osuji and Onojake, 2004, 2006; Nie *et al.*, 2010).

Previous researchers have investigated the simultaneous contaminations of heavy metals and oil. The oil spillage was partially responsible for the concentrations of Ni, Cu and Pb in oil-polluted soils in Nigeria (Onojake and Frank, 2013). No significant metal (As, Cd, Cu, Hg, Ni, Pb, V) enrichment is detected from the *in-situ* oil sand industry at the Cold Lake oil field (Skierszkan *et al.*, 2013). Some researchers believed that the crude oil exploitation activities (including heavy metal pollutions) negatively impacted human health in northeast British Columbia (Krzyzanowski, 2012). However, little research has focused on the distribution and potential sources in oil-polluted soils in oilfields of China, where the soil texture and the components of crude oil are different from those of other countries. The Yellow River Delta is a riverine delta in Shandong Province, China, where the Shengli Oilfield

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*²Corresponding author. E-mail: cuiwj@sdu.edu.cn.

is located and has been exploited since the year 1961. Petroleum hydrocarbon contamination in this region is severe, so the associated distribution and sources of heavy metals in the soil of this region must be different from those of the areas without oil pollution in China.

In this study, a typical crude oil-polluted region in the Yellow River Delta, China was chosen to study the spatial distribution and potential sources of heavy metals in oil-polluted soils.

MATERIALS AND METHODS

Study area and soil sampling

To study spatial distributions and sources of heavy metals in oil-polluted soils, Gudao town was chosen. Gudao is one of the most important regions of the Shengli Oilfield, the second biggest oilfield in China. Gudao town is a significant new oil-industry town that developed based on oil exploitation. The typical characteristics of the town are a long oil exploitation history (since 1970) and a short urbanization history (since 1992). The total area of Gudao is about 160 km², and the population is about 37 000. It is in the East Asian monsoon region. The average annual temperature is 12.1 °C and the average annual precipitation is 580 mm. The soils in this area are mainly fluvo-aquic soils and coastal solonchaks (World Reference Base for Soil Resources).

In this study, 22 surface soil samples (0–20 cm depth) (Fig. 1) were collected in April, 2012. The study area (sub-regional scale, 50 km²) was located within latitudes 37°50'0" and 37°55'0" N, and longitudes

118°44'0" and 118°49'0" E. Samples were taken based on a grid system divided into 2-km² squares; however, when the access was too difficult to obtain, samples were not taken at grid points. For each soil sample, five subsamples with similar weight were collected using a plastic scoop and a stainless steel shovel. Then, the subsamples were mixed together and the sample weight was decreased to about 1 kg using the quarter method (SEPAC, 2004) to improve the sample representativeness. Each soil sample was stored in a plastic bag. The coordinates of the sampling locations were recorded simultaneously by a GPS (eTrex Ventruer, Garmin Corporation, China).

Chemical analysis

The organic matter (OM) content of the soil samples was determined by the Walkley and Black method (Walkley and Black, 1934). The soil pH was determined at a soil:water ratio of 1:5 with a pH meter. Salinity was measured in a deionized suspension (1:10, w/w) with a standard conductivity meter (DDS-11A, Shanghai Precision Scientific Instrument Co., Ltd. (INESA), Shanghai, China) (Nie *et al.*, 2010). The concentrations of total petroleum hydrocarbons (TPHs) of the soil samples were determined by gravimetric analysis (Adeniyi and Afolabi, 2002).

To determine the concentrations of total heavy metals, the soil samples were dried at room temperature, and then ground to pass through a 100-mesh nylon sieve after large impurities (such as plant roots and stones) were discarded. Each soil sample (about 0.500 g) and 10 mL of HCl were transferred into a polytetrafluoroethylene (PTFE) crucible. The crucible was placed overnight (12 h) to oxidize organic matter preliminarily. Then the solution was heated on an electric hot plate at about 95 °C to reduce the volume to about 3 mL. After cooling, 5 mL HNO₃, 5 mL HF and 3 mL HClO₄ were added, and the crucible was covered and heated at about 120 °C for 2 h to destroy organic matter and silicates of the soil samples. Then the crucible was uncovered and heated continuously until the solution was nearly dry and fumes nearly disappeared. The residue should be a viscous fluid which is colorless or light yellow. If the residue was still dark, a further 5 mL HNO₃ was added and the heating process was repeated until the residue met the color requirement. After adding 2.5 mL HNO₃ and 10 mL deionized water, the crucible was then gently heated to dissolve the residue. The final solution was transferred into a volumetric flask and made up to 50 mL with deionized water. In the whole process, the HCl, HNO₃, HF and HClO₄ used were all concentrated acids. Concentrations

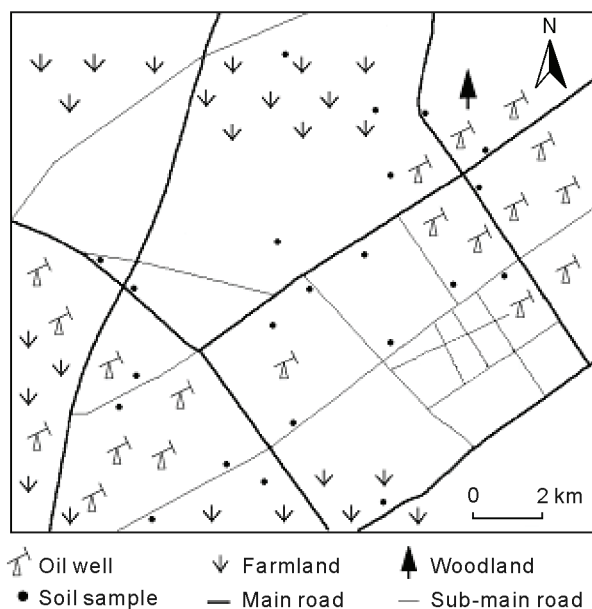


Fig. 1 Soil sampling locations in the study area (Gudao Town, Shandong Province) in the Yellow River Delta of China.

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