

Effects of Crude Oil Contamination on Soil Physical and Chemical Properties in Momoge Wetland of China

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Abstract: Large oilfields are often coincidentally located in major river deltas and wetlands, and potentially damage the structure, function and ecosystem service values of wetlands during oil exploration. In the present study, the effects of crude oil contamination during oil exploration on soil physical and chemical properties were investigated in marshes of the Momoge National Nature Reserve in Jilin Province, China. The concentrations of total petroleum hydrocarbons in the marsh soil near the oil wells are significantly higher than those in the adjacent control marsh. Soil water contents in oil-contaminated marshes are negatively correlated with soil temperature and are significantly lower than those in the control area, especially in fall. Crude oil contamination significantly increases the soil pH up to 8.0, and reduces available phosphorus concentrations in the soil. The concentrations of total organic carbon are significantly different among sampling sites. Therefore, crude oil contamination could potentially alkalize marsh soils, adversely affect soil fertility and physical properties, and cause deterioration of the marshes in the Momoge National Nature Reserve. Phyto-remediation by planting *Calamagrostis angustifolia* has the potential to simultaneously restore and remediate the petroleum hydrocarbon-contaminated wetlands. Crude oil contamination affects the soil physical and chemical properties, so developing an effective restoration program in the Momoge wetland is necessary.

Keywords: crude oil contamination; marsh soil; oilfield in marshes; soil physical and chemical properties; wetland; Momoge National Nature Reserve

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1 Introduction

Oil contamination can affect soil physical and chemical properties. The daily maximum surface temperature of hydrocarbon-contaminated soils is often higher than that of adjacent control sites (Aislabie *et al.*, 2004). Oil usually causes anaerobic environment in soil by smothering

soil particles and blocking air diffusion in the soil pores, and affects soil microbial communities (Townsend *et al.*, 2003; Labud *et al.*, 2007; Sutton *et al.*, 2013). Heavy crude oil pollution can cause complete mortality of marsh vegetation (Lin and Mendelssohn, 1996; 2012). In addition, crude oil-contaminated soils are hydrophobic compared with pristine sites (Quyum *et al.*, 2002;

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Aislabie *et al.*, 2004). Hydrocarbon contamination can also increase soil total organic carbon (Ekundayo and Obuekwe, 2000), and change soil pH values (Hu *et al.*, 2006; Wang *et al.*, 2009; 2010) and other soil chemical properties (Arocena and Rutherford, 2005; Kisic *et al.*, 2009).

The Momoge National Natural Reserve (MNNR), located in Zhenlai County, Jilin Province, China, is one of the largest inland wetland and hydrosphere ecosystems in China (Xu and Gu, 2003). There are abundant crude oil resources beneath the MNNR, geographically coincident with the wetlands, which have huge strategic and economic importance and value. The MNNR is in a temperate continental monsoon climate zone with an average annual temperature of 4.2°C, average temperature of 23.5°C in July, and an annual precipitation of 380–412 mm. During flooding seasons or rainy years, the Nenjiang River usually backflows and the Tao'er River overflows. Thus, surface runoff accumulates and forms a pool swamp wetland. The MNNR includes most wetland classification types in the Songnen Plain, such as swamps, lakes, salt marshes, paddy fields, and others. Wetland ecosystems, as the main protected areas in the reserve, have unique ecological values and functions, especially for rare birds and waterfowl, such as the red-crowned crane, which inhabit the wetlands (Yuan *et al.*, 2005).

Healthy soil environments are not only important structures and functions of wetland ecosystems, but also are important for the health of humans, animals and plants. However, since 1986, large scale oil exploration has occurred in the core area of the MNNR, and severely threatened wetland ecosystems, resulting in a reduced wetland area, a decrease in the dominant sedge (*Carex tato*) and narrow-leaf small reed (*Calamagrostis angustifolia*) plants, as well as decreased and damaged rare and endangered waterfowl habitats. The core zone and buffer zone of the MNNR occupy 36% and 22% of the total area of the MNNR, respectively. The experimental zone for crude oil exploration occupies 25% of the area of MNNR (Yuan *et al.*, 2005). Several previous studies have been conducted on the MNNR (Wang *et al.*, 2009; 2010; Bai *et al.*, 2012), however, little attention has been paid to the seasonal effect of crude oil pollution on the local wetlands.

In this study, we investigated the seasonal effects of crude oil pollution on some major soil physical and

chemical properties in MNNR. The properties investigated included concentrations of the total petroleum hydrocarbons (TPH) in the marsh soils, total organic carbons (TOC), available phosphorus (AP), pH, soil moisture and soil temperature. The results of the present study will provide information for potentially remediating and restoring crude oil-impacted marsh soil ecosystems in the MNNR and other similar wetlands.

2 Materials and Methods

Five marsh soil sampling sites (45°52'06"–45°53'54"N, 123°57'30"–123°59'42"E) with different TPH contents and restoration treatments were selected from the MNNR (Fig. 1). Among these five sampling sites, the control site was in an un-quarried area (in the buffer area of the MNNR without oil contamination, near the experimental area and oilfield exploration area), while the other four sites were located in the oilfield exploration area, including the sites of oil-1 and oil-2 (without any vegetation), and the sites of oil-1-planting and oil-2-planting which had been remediated by planting the local dominant plants narrow-leaf small reed (*C. angustifolia*) for one year earlier. All the four sites had been explored for oil over 10 years previously. The oil-1 site was close to the oil-1-planting site, and the oil-2 site was adjacent to the oil-2-planting site.

There were five replicates for each sampling site. For the polluted sites, the soils were collected about 2 m away from their respective abandoned oil wells. To avoid potential difference among sampling sites arising from different sampling time, we collected all the samples between 1:00 pm and 2:00 pm on the same day. Soil samples were taken three times in May, August and October of 2009.

In the field, soil temperature was measured by inserting a thermometer about 5 cm deep into the soil. After removing the back-filled clean soil covering on the surface, we collected soil samples at 5 cm below the surface, and analyzed various properties in the laboratory within 24 h. The collected samples were analyzed for soil moisture by the oven drying method (Wilke, 2005), soil TPH content by a gravimetric method (Mishra *et al.*, 2004), pH with a pH meter (Delta 326, Shanghai) (Wilke, 2005), AP as sodium bicarbonate (NaHCO₃)-extractable phosphorus (Schoenau and O'Halloran, 2008), and TOC by the potassium dichromate dilution

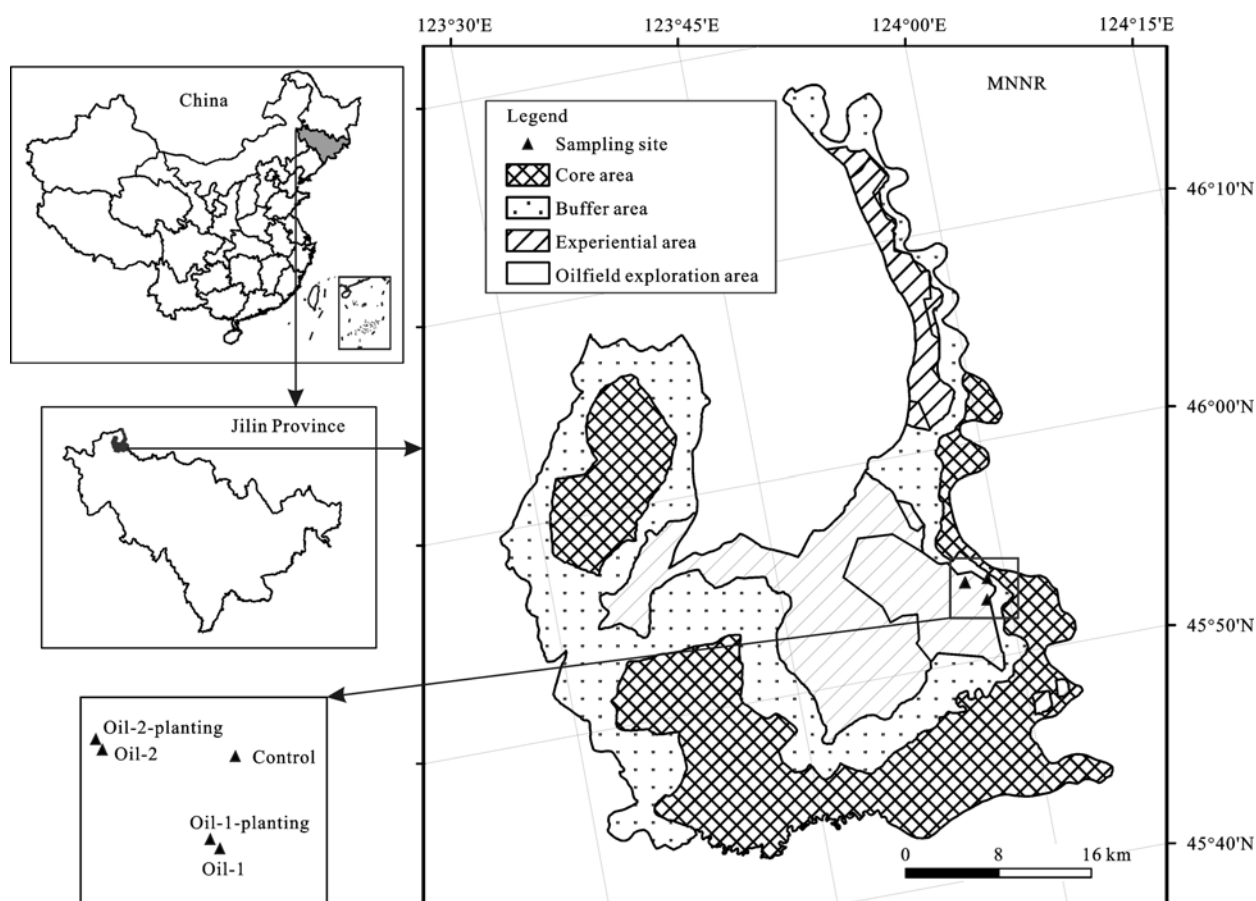


Fig. 1 Location of sampling sites in Momoge National Natural Reserve (MNNR)

heat colorimetric method (Bao, 1999).

Statistical analysis was conducted by using the Statistical Analysis System (SAS) with general linear models (Proc GLM). Least squares means were used to evaluate statistical differences among means of the main factors and individual treatment-level combinations. Significant differences are reported at the 0.05 probability level unless otherwise stated.

3 Results and Analyses

3.1 Effect of crude oil pollution on total petroleum hydrocarbons in soil

The concentrations of TPH of the marsh soil in the oil-field sampling sites were significantly higher than those of the control soil (Fig. 2). The TPH concentrations in oil-1-planting and oil-2-planting sites were substantially lower than those in the oil-1 and oil-2 sites, respectively, which indicated that phyto-remediation by planting *C. angustifolia* enhanced oil attenuation.

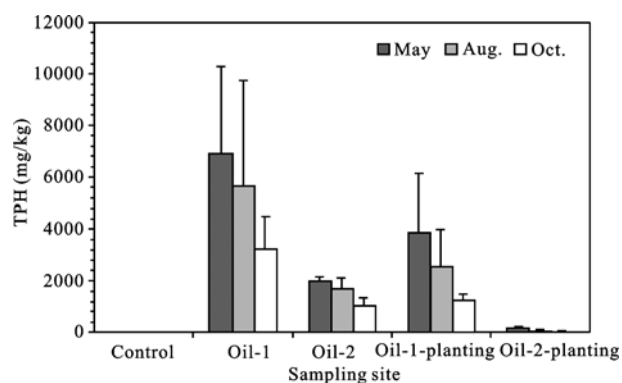


Fig. 2 Changes of total petroleum hydrocarbon (TPH) concentrations in different sampling sites and seasons. Values are means with standard errors ($n = 5$)

The oil attenuation rate of 74% at the two oil-planting sites tended to be higher than the rate of 50% at the oil-1 and oil-2 sites during the period of May–October, although the difference was not statistically significant because of the short duration of the phyto-remediation period. These results may suggest that planting the local

dominant wetland plant *C. angustifolia* simultaneously restores oil-damaged wetlands and prevents degradation of crude oil in wetland soils. Phyto-remediation is considered to increase oil attenuation by the plant taking in small molecular hydrocarbons. Aerating the soil to increase aerobic degradation of oil provides root exudates for microbial co-metabolization of oil components and other molecules (Lin and Mendelsohn, 2009). The oil content in the oil-2-planting site was almost reduced to the level of the control.

The marsh soil also showed a natural self-purification capability in that crude oil in the same sites even without planting decreased with time, most likely because of soil microbial degradation and oil weathering (Mendelsohn *et al.*, 2012). By comparison with the TPH concentrations in a previous study (Wang *et al.*, 2010), the TPH concentrations in the present study were generally substantially lower than those determined in July 2007, suggesting that microbial degradation, phyto-remediation, and oil weathering processes could play important roles in oil attenuation in the Momoge wetlands over long periods.

3.2 Effect of crude oil pollution on temperature and water content of soil

The concentrations of TPH in crude oil significantly affected soil temperature ($p < 0.0001$). Soil temperature also significantly fluctuated with seasons ($p < 0.0001$), and an additionally significant interaction ($p = 0.006$) between the concentrations of TPH in crude oil and sampling seasons suggested that the effect of oil pollution on soil temperature changed with season. In May, soil temperature in all oil-contaminated sites was significantly lower than that in the control site (Fig. 3). In August, there were no significant differences among all sites. However, in October, the soil temperature in the two planted sites was significantly lower than that in the control site. The soil temperature was the highest in the summer, and the lowest in the fall when the temperature dropped quickly below 10°C in October.

Crude oil pollution significantly affected soil water contents. Soil water content also significantly fluctuated with seasons ($p < 0.0001$), and an additionally significant interaction between the concentrations of TPH in crude oil and sampling seasons ($p = 0.0004$) indicated that the effect of oil pollution on soil water content also changed with season. The soil water content was the

lowest in summer ($7.8\% \pm 0.9$, $n = 25$), and the highest in fall ($23.7\% \pm 1.1$, $n = 25$), while the values in spring fell between the two ($16.8\% \pm 1.8$, $n = 25$). In October, the soil water contents at all oil-contaminated sites were significantly lower than that at the control site (Fig. 4). Hydrophobic crude oil coats soil particles, blocks soil pores, and reduces the permeability of water and air (Bennett *et al.*, 1993; Roy *et al.*, 1999; Khamehchiyan *et al.*, 2007), and thus affecting the soil water content.

However, in May and August, soil water contents were not significantly different among all sites except for the oil-1 site in May, which was probably caused by the variation of soil temperature. The changes in marsh soil water content were negatively correlated with soil temperature ($r = -0.789$) (Fig. 5), suggesting that about 70% of the soil water content may be explained by soil temperature. Other factors controlling soil water content may include precipitation, evaporation and marsh plant

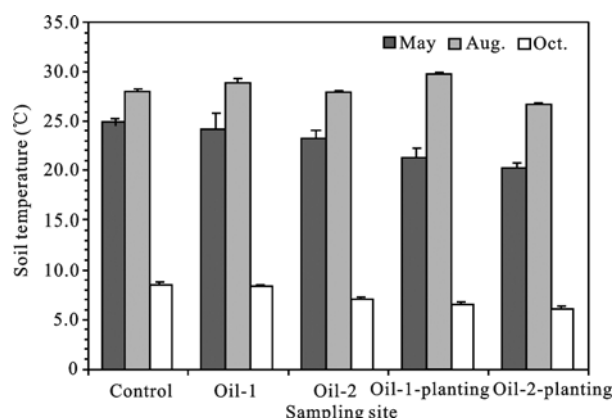


Fig. 3 Changes of soil temperature in different sampling sites and seasons. Values are means with standard errors ($n = 5$)

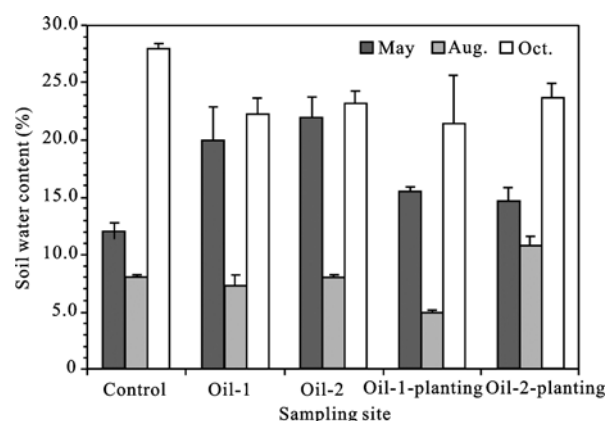


Fig. 4 Changes of soil water content in different sampling sites and seasons. Values are means with standard errors ($n = 5$)

transpiration, although direct water supply is probably one of the most important factors. The significant seasonal changes in the soil temperature are considered to be the most likely driver of the fluctuations in soil moisture. High soil temperature generally induces high water evaporation from soil and water surface, while the concurrent presence of substantial live vegetation biomass with high temperature in summer may cause additional high vegetative transpiration from plant leaf surface. The combination of high physical evaporation and plant transpiration potentially resulted in low soil moisture in summer.

The study area is located in the northern temperate and semi-arid region. Human activities and global warming have been increasingly affecting the water supply of the region, although the human influence is less compared with the impact of natural factors (Jiang *et al.*, 2007). Therefore, oil exploration might exaggerate drought conditions in the wetland, especially in the dry years, thereby potentially further increasing damage to the wetlands resulting from alkalescence in this semi-arid region. The results of this study showed that crude oil pollution in the abandoned oil field led to lower soil water content, indicating that the oil pollution might suppress water use efficiency in the Momoge wetlands.

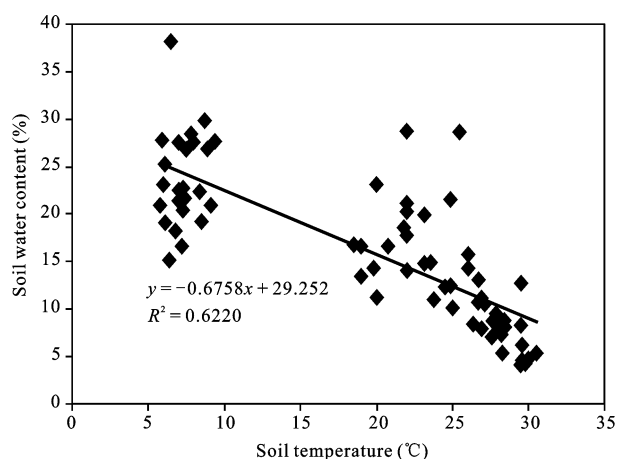


Fig. 5 Relationship between soil water content and soil temperature from May to October

3.3 Effect of crude oil pollution on pH of soil

Crude oil pollution increased pH values in the marsh soil. The pH values in each sampling site showed no significant difference among seasons, so the concentra-

tions were averaged over the sampling seasons to show the effects of oil contamination (Fig. 6). The soil pH values in all the oilfield sites were up to 8.0 and were significantly higher than that in the control site ($p < 0.0001$) except that in the oil-2 sampling site (Fig. 6). There was no significant effect on the soil pH values among seasons ($p > 0.05$) which differ from the results of previous studies that an increase of TPH concentrations in soil could lower pH values (Leahy and Colwell, 1990; Gong *et al.*, 2008; Kistic *et al.*, 2009).

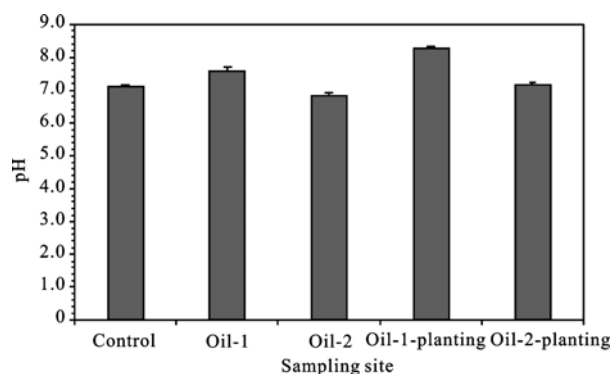


Fig. 6 Changes of soil pH at different sampling sites. Values are means averaged over seasons with standard errors ($n = 15$)

The results of previous studies on oilfields in China showed that oil pollution raised soil pH (Jia *et al.*, 2009; Wang *et al.*, 2010), which is supported by our results. The higher pH values in crude oil-polluted soil in this study might be caused by two factors: first, the hydrophobic nature of crude oil might induce a potential drought in the surface and subsurface layers of polluted soil (Njoku *et al.*, 2009), which could aggravate salinization, and thus raise the pH values compared with that in the control site; second, oil pollution in soil has been shown to be associated with the accumulation of exchangeable base (such as Ca^{2+} , Na^{+}) and a reduction in exchangeable acidity and effective cation exchange capacity (ECEC) (Benka-Coker and Ekundayo, 1995; Ekundayo and Obuekwe, 1997; Osuji *et al.*, 2006; Agbogidi *et al.*, 2007). These mechanisms might also underpin the increase of pH values in the crude oil polluted soil.

Crude oil elevating pH in the soil could potentially exaggerate the damage attributable to alkalescence in the region, which might threaten the wetland values of the MNRR. Whether the increase of soil pH caused by crude oil pollution in the Momoge wetlands is affected

by geological location and the specific type of crude oil will require further investigation.

3.4 Effect of crude oil pollution on available phosphorus of soil

Crude oil in the oilfield marshes reduced AP concentration in the soil. The concentrations of AP in the soil of all oilfield sites were significantly lower than that in the control site ($p < 0.0001$) (Fig. 7). In addition, the AP concentrations were significantly different among the sampling seasons, being 13.9 ± 2.8 mg/kg ($n = 25$) in May, 22.7 ± 3.7 mg/kg ($n = 25$) in August and 18.4 ± 3.1 mg/kg ($n = 25$) in October. The AP concentration in each sampling site showed no significant difference among seasons ($p > 0.05$), so the concentrations were averaged over the sampling seasons (Fig. 7) to show the effects of oil contamination.

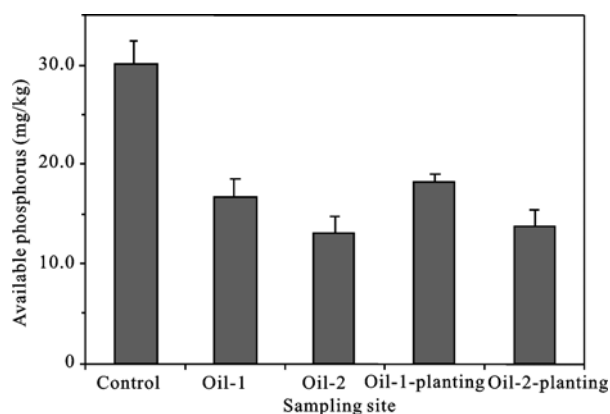


Fig. 7 Changes in available phosphorus (AP) concentrations of soil at different sampling sites. Values are means averaged over seasons with standard errors ($n = 15$)

The results of previous studies showed that oil contamination decreased AP concentration by various degrees (Wang *et al.*, 2009; 2010; Eneje *et al.*, 2012). A field study on the Momoge wetlands showed that the concentration of AP decreased with increasing time of oil exploration and production (Wang *et al.*, 2010). In an experimental oil study, the concentration of AP in the crude oil-contaminated soil decreased as much as 66% compared with the control site when the content of crude oil reached 30 mg/kg (Eneje *et al.*, 2012). However, Liu *et al.* (2007) reported that AP concentration was not significantly affected by oil contamination.

In the present study, lower AP concentrations in all oilfield sites than in the control site, may be caused by

two reasons. First, TPH in the soil could increase the carbon concentration, which might affect the equilibrium of nutrients in the soil. Microbes in soils, which utilize TPH as a carbon source, could utilize considerable amounts of AP when they degrade the hydrocarbons (Jobson *et al.*, 1974; Braddock *et al.*, 1997; Wang *et al.*, 2009). Second, phosphorus solubility is maximized at pH 6.5 (Riser-Roberts, 1998), and consequently the higher pH values in oilfield sites (Fig. 6) could also lower the AP concentration compared with the concentration in the control site (Fig. 7). Phosphorus is one of the most important macro-nutrients for plants and soil microorganisms. The decrease of AP concentrations in oilfield marshes could change the structure of vegetation and soil microorganisms, and reduce marsh ecosystem services and values.

3.5 Effect of crude oil pollution on total organic carbon of soil

The concentrations of TOC were significantly different among sampling sites ($p < 0.0001$). However, sampling seasons did not significantly affect the TOC concentrations, and hence the concentrations were averaged over the sampling seasons (Fig. 8).

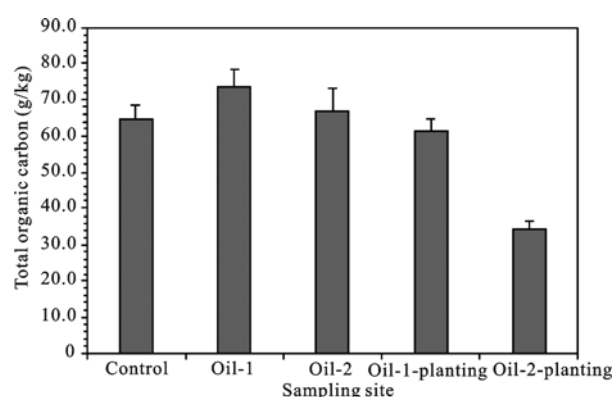


Fig. 8 Changes in total organic carbon (TOC) concentrations of soil at different sampling sites. Values are means averaged over seasons with standard errors ($n = 15$)

Organic carbon in the soil is generally derived from biota, such as peat formation with time, plant fine roots turnover, microbial biomass and others. However, crude oil contamination in oilfield marshes might also contribute to the TOC in the soil. The low concentration of TPH in the oil-2-planting site might result in a lower TOC content. However, in this study, the organic carbon derived from petroleum hydrocarbons contributed less

than 10% of the TOC, because the average TPH concentration in the oil-contaminated marshes was less than 0.3% compared with an average TOC concentration of about 6%. Wang *et al.* (2009; 2010) reported that oil contamination significantly increased the TOC contents most probably because of the much higher TPH concentration (up to 3%) in their study than that in this study.

4 Conclusions

Crude oil pollution detrimentally affect soil physical and chemical properties of the oilfield marshes in the MNNR. The presence of oil in the soil lower soil fertility by reducing AP, and increasing soil pH, which potentially accelerated damage attributable to alkalescence in the wetlands in this semi-arid region. In addition, oil pollution might adversely affect the marsh water use efficiency by lowering the soil water content, which could be especially critical in the MNNR where the semi-arid conditions lead to the water supply being relatively limited. The results of phyto-remediation by planting *C. angustifolia* show the potential of simultaneously restoring and remediating the petroleum hydrocarbons-contaminated wetlands, but most likely need longer time before it effectively improves the physical-chemical characteristics of petroleum-contaminated soils, and contributes to oil degradation of the crude oil exploration-damaged marsh soil ecosystem in the MNNR. How crude oil contamination affects the interactions between soil chemical factors has not been resolved in this study, which will be studied in the future.

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