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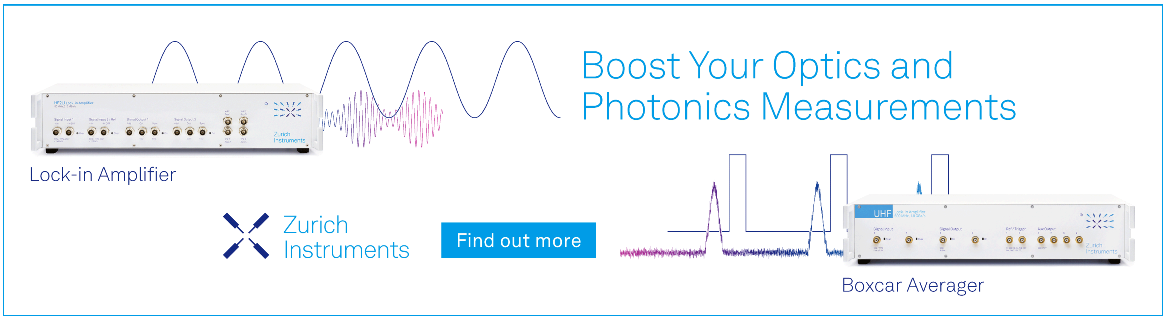

The Harm of Petroleum-Polluted Soil and its Remediation Research

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Abstract. Land resources are the foundation of human's survival and development, and it's one of the most valuable natural resources of each country. In view of the serious problems of petroleum pollution to soil caused during the exploration and development processes, this article based on a large number of literature researches, firstly discussed the compositions and properties of petroleum contaminants, secondly investigated some restoration methods for the current situation of petroleum polluted soil, compared and analyzed the advantages and disadvantages of three kinds of bioremediation technologies. Finally, according to the deficiencies of previous research and existing problems, made an outlook of the physical and chemical remediation, bioremediation, and microbe-plant remediation, to provide some enlightenments for petroleum-contaminated soil remediation.

Key words: petroleum pollution; soil; contaminations; remediation technologies.

INTRODUCTION

Soil is the material basis for the sustainable economic and social development, and is one of the most valuable natural resources in each country, especially for our country. With continuous improvements of China's industrialization and urbanization level, the demand for oil products rises sharply (Tang, 2014). Now in China a total of more than 400 oil and gas fields distributed in 25 provinces and cities and autonomous regions nationwide. The main scope of oil fields is nearly 200000 km^2, covering an area of 320000 km^2, which accounted for nearly 3% of the total area. Among them, the oil content in the soil of nearly 4.8 million hm^2 may be over the secure value (Liu et al, 2007). During the process of oil and gas exploration, development, gathering, refining, storage and sales, the accidents, incorrect operations and equipment maintenances and other reasons would result in the overflow and emissions of petroleum hydrocarbons (Shi et al, 2013).

According to some related statistics, China's oilfield enterprises produce 7 million tons ground crude oil per year, and the ground crude oil of single well can be up to about 2 tons (Du et al, 2011). In addition, for the past few decades the production technologies of most oil fields are relatively low (Zhu et al, 2013), the corresponding environmental protection measures and evaluation systems are imperfect, out-dated pollution control and remediation technologies lead to lots of serious soil pollutions. Recently according to the deployment of the Party Central Committee and the State Council, we should actively promote the protection and restoration of mountains-forests-lakes ecosystem; strengthen the land management and pollution remediation. In view of this, this article will discuss the harm of petroleum-polluted soil and repair management, to provide some good ideas for petroleum-contaminated soil restoration.

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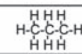
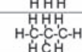
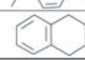
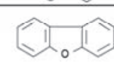
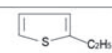
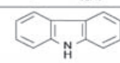
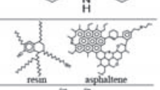
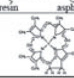
THE COMPOSITION AND PROPERTIES OF PETROLEUM CONTAMINANTS

The petroleum is mainly composed of various hydrocarbon complex mixtures. It can be divided into saturated hydrocarbon, aromatic hydrocarbon and non-hydrocarbon compounds (Table 1) by chromatography (Liu et al, 2009; Lu and Zhang, 2008). The saturated hydrocarbon molecular structure consists of carbon-carbon bond and carbon-hydrogen bond, which is easy to be degraded. In addition, its boiling point is relatively low, so the saturated hydrocarbon can gradually disappear from the soil in the way of photosynthesis and volatilization. However, the molecular structure of aromatic hydrocarbon is quite complex. The complex benzene ring and its higher boiling point greatly increase the difficulties of being removed from the soil. The polycyclic aromatic hydrocarbon is the typical POPs that widely exists in various environmental systems (Guo et al, 2016; Bayat, 2015; Li, 2008), and the 16 kinds of PAHs have been included in the list of priority control pollutants by EPA of the United States and European Community (Wan, 2014; Binet et al, 2000), namely naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, diphenyl[a,h]anthracene, benzene [ghi]perylene and indene and [1,2,3-cd] pyrene.

TABLE 1 The composition of petroleum group

petroleum	group	composition	molecular structure	boiling point	the degradation order
hydrocarbons	saturated hydrocarbons	n-alkanes（straight chain）		lower	straight-chain paraffin > branched paraffin > cycloalkanes > naphthenoaromatic hydrocarbon > aromatic hydrocarbon > non-hydrocarbon
		isoparaffin（branched chain）			
		cycloparaffin			
	unsaturated hydrocarbons	aromatic（monocycle、bicyclo、polycyclic）		high	
		naphthenoaromatic hydrocarbon			
non-hydrocarbons	oxy-compound	acids: saturated acid、naphthenic acid、isoamyl diene between acid、aromatic acid		higher	
	sulfo-compound	mercaptan、monosulfide、disulphide、thiophene derivative			
	n-containing compound	pyridine、quinpline			
	resin and asphaltene	polycyclic aromatic nucleus or naphthenic aromatic nucleus			
	organometallic compound	porphyrin-like compounds			

Among the three kinds of hydrocarbon compounds, non-hydrocarbon compounds have the maximum carbon number. They are insoluble in water, and the fusion and boiling point are quite high, so they are most difficult to be removed from the soil, resulting in obvious environmental toxicity and mutagenicity (Liu, 2012; Chalneau and Morel, 1997). Therefore, the natural soil residual poison composition is mainly polycyclic aromatic hydrocarbons and non-hydrocarbon compounds. In addition, table 1 shows that non-hydrocarbon composition contains a small amount (usually ppb~ppm magnitude) of heavy metal elements such as vanadium and nickel (Liu and Zhang, 1991; Carls et al, 1995) and some studies have reported that the drilling fluid also contains heavy metal pollutants, with high toxicity, persistence, and refractory (Fu,2014; Kisic et al, 2009).

Besides oil, the sewage in oil and gas fields can also lead to soil pollution. At present, most domestic oil fields have entered into the middle and later periods of oil production, water content of the crude oil reaches to 70%~80%, some even up to 90%. Large amounts of oily sewage will produce after oil/water separation. If untreated, they could lead to serious soil and water pollution (Lu, 2009; Gu et al, 2007; Mariana et al, 2010). In terms of its composition, the oilfield sewage contains oil, various salts, organic matter, inorganic matter and some microbes etc (Table 2). The salinity is higher, generally between 10^3~14×10^4mg/L, the main salts are Na_2SO4, $NaHCO_3$, $MgCl_2$ and NaCl (Liu, 2009; Li, 2010). This kind of waste water not only causes soil salinization, but also destroys the soil environment quality.

TABLE 2 The composition of oilfield sewage

type	primary materials
crude oil	oil content 1000-2000mg/L， oil spill、dispersed oil、emulsified oil and dissolved oil
inorganic salts	Ca^{2+}、Mg^{2+}、K^{+}、Na^{+}、Cl^{-} 、HCO_3^{-}、CO^{2-}、SO_4^{2-}
organics	aliphatic hydrocarbon、aromatic hydrocarbon、phenols、organic sulfide、aliphatic acid、polymers
inorganic matter	H_2S、FeS、clay particles、silt and fine sand
microorganism	sulfate reducing bacteria、saprophytic bacteria and iron bacteria

THE HARM OF OIL-POLLUTED SOIL

Through a large number of literature research, the harm of oil-polluted soil mainly includes the following aspects:

Firstly, because of the small density, higher viscosity and lower emulsifying ability of petroleum, it is easy to be absorbed in soil surface, affecting the permeability and porosity of soil (Wang, 2009; He et al, 1999); petroleum is rich in carbon and a small amount of nitrogen compounds, so it can change the composition and structure of soil organic matter and impact the C/N, C/P, salinity, pH, EH and conductivity of soil (Li et al, 2009). The heavy metals (nickel and vanadium) in oil mixtures (Saadat et al, 2014) and high concentrations of salt in oilfield output water can also damage the soil environment (Efsun et al, 2015).

Secondly, microorganisms in natural environment are quite abundant in healthy and clean soil. In normal situation the microorganisms which can resist the oil pollution stress are not developed, while in contaminated soil, in order to adapt to this kind of environment, they can produce certain enzyme system and gradually form a dominant population with symbiotic or synergy effect (Chiara et al, 2009). A number of studies have shown that the hydrocarbon pollution can change the microbial population, the composition of the community structure and the enzyme system in soil, given priority to the inhibitory action (Deng, 2014; Uzoije and Agunwamba et al, 2009).

Thirdly, it can impede the normal growth of crops such as reduce the germination rate and fertility and decline the resistance to pests and diseases (Xu and Lu, 2010; Zhu, 2010; Shan et al, 2014). In addition, the oil compounds could react with inorganic nitrogen and phosphorus, limiting the nitrification and removal of phosphoric acid, so the effective nitrogen and phosphorus in the soil would decrease and the absorption of crops will be affected (Liao et al, 2015; Pinchin et al, 2013; Shen, 2011).

Moreover, the polycyclic aromatic hydrocarbons in petroleum chemicals have carcinogenic, mutagenic, teratogenic and other toxic effects. It can enter into the bodies of people and animals through breathing, skin contact and diet, degrading the normal function of livers and kidney etc, therefore causing great threat to human's health.

At last, the oil pollutants in the soil not only impact the pedosphere, but also the atmosphere and water sphere. To be specific, the low boiling point and light weight hydrocarbons can enter into the atmosphere by evaporation easily; then through runoff and infiltration into the surface water and osmosis into the groundwater system; and finally through the food chain enter into the human's bodies (Zhang, 2006).

OIL POLLUTION REMEDIATION RESEARCH STATUSES AT HOME AND ABROAD

Recent years, many countries in the world have successively conducted many researches on the restoration of oil polluted soil. The common methods are physical, chemistry, microbiology and plant remediation (Han, 2013). The traditional physical and chemistry method, which include soil removal and replacement, elution method, heat treatment and thermal resolution, extraction-separation and chemical oxidation methods, are used to regulate the physical processes or chemical reactions to change the physical properties of the soil and then control the contaminants effectively. This method is generally more thoroughly and stable, while it also need to build some fixed processing facilities, use chemical agents; and during the operation the temperature, pressure and power supply are required to handle the whole process (Han, 2013; Zhang et al, 2001; Thapa et al, 2012). In view of its high cost, secondary pollution and not suitable for large area remediation, it is not widely used in actual application.

By contrast, the bioremediation technology has many benefits such as low cost, simple in-situ treatment, environmental friendly, no secondary pollution, and removes some pollutants with high efficiency (Hill et al, 2000; Shen et al, 2002; Liste and Alexander, 2000), attracting a lot of domestic and foreign scholars. According to the type of organism, the bioremediation technology generally can be divided into three categories: microbial remediation, phytoremediation and plant-microbial technology.

Since 1965 Jones and Smith firstly made use of microbes to dispose of the petroleum, the micro biodegradation method was created (Tang, 2013). At present there are more than 100 species, 200 species of microorganisms that can degrade the oil pollutants. Based on the source of microorganisms, they can be divided into indigenous microbes, alien microbes and genetic engineering bacteria. Of these three organisms, the indigenous microorganisms are able to adapt to the oil pollution and keep higher microbial activity and degradation efficiency, so they are the most widely applied in the microbial remediation (Yao et al, 2006; Yang et al, 2008; Kuiper et al; 2004), but naturally the number of indigenous microbial can't meet the demand of reparation, in practice, the indigenous degrading bacteria was isolated from the contaminated soil, after expanding cultivation then poured into the soil to improve the efficiency of degradation (Tian, 2013; Peng, 2009).

Phytoremediation is primarily using the interactions between plants, microbes and environment to form a soil-plant-rhizosphere microorganism multiple system. It's like a powerful "live purifier", including the solar-powered "water pumps", "plant reactor", "microbial converter" and "soil filter" (Wang et al, 2008; Cheng et al, 2003; Mohn et al, 2001). The main three mechanisms to remove oil contaminates are direct absorption, releasing secretions and enzymes, stimulating microbial activity in the root area and strengthening biological transformation. Studies have shown that the removal of most pollutants in the plant’s root soil is mainly due to the degradation or transformation of microorganisms. On one hand, the secretion released from the root system causes the bacterial enzyme system to secrete the degrading enzyme, then directly stimulate the growth of degrading bacteria. On the other hand, the root system grows and promotes the soil to form an effective water channel, which improves the permeability of the soil (Child et al, 2007; Josiah, 2009; Roling, 2003).

In reality, because of the complex composition of petroleum pollutants, a single repair method often does not work well. While the microbe-plant reparation is to use the complex system of soil-plant-microorganisms to remove the pollutants, so it can degrade and remove the pollutants more efficiently. For the interaction of microbes and plants often occurs in the root system, the key to this technology is the rhizosphere regulation of oil pollutants. As for this method, the plants provide nutrients for microorganisms through photosynthesis and casts, which contain sugar, alcohol, proteins and organic acids, etc. The root secretion promotes the degradation of oil pollutants by microorganisms. At the same time, the root extension not only provides the living place for the degrading bacteria, also promotes the aerobic function of the root system smoothly.

In recent years, scholars at home and abroad have got some achievements on microbe-plant reparation technology. Child (2009) used five kinds of PAHs-degrading bacterium to impregnate the barley seeds. He discovered that the mycobacterium directly attached to the root system by seed germination, improving the degradation rate of PAHs in the soil. Joner (2003) grew rye grass and white clover in two different concentrations of PAHs, after 28 weeks it was found that the root biomass of arbuscular mycorrhizal fungi (AMF) system was 1.65 times than before. The larger root biomass and surface area are beneficial to the adsorption and absorption of organic matter. Domestically, Gao (2013) made use of the combination of high efficient petroleum degradation bacteria and plants to repair the oil-heavy metal pollution in Gudao oil province. This research had shown that the petroleum hydrocarbon degradation rate of joint repair technology is significantly higher than that of single repair technology. Li (2013) used suaeda, the local salt raw plant, and dominant mycorrhizal fungi in Dongying region to establish the AMF-plant-nitrogen addition joint repair system. Through container experiments she found that the oil degradation rates of AMF-Suaeda and Carbamide-AMF-Suaeda increased significantly. To sum up, the microbe-plant combination system can obviously improve the removal efficiency of petroleum pollutants in the soil. The following are advantages and limitations of three kinds of remediation technologies, shown in table 3.

TABLE 3 The benefits and limitations of microbes, plants and plant-microbial remediation technologies

type	benefits	limitations
microbial remediation	in-situ or ex-situ remediation, low cost, diversified practices	microbial activity is easy to be affected by temperature, oxygen, water and Ph etc
phytoremediation	in-situ remediation, low cost, simple operation, purify the air and water, suitable for large-area and low concentration polluted soil, without space limitation and has aesthetic value	long period and inefficiency, some plants are sensitive to soil fertility, climate, water, salinity, Ph, palnt diseases and insect pests
plant-microbe remedaition	in-situ remediation, efficiently, suitable for some persistent organic pollutants, and the mycorrhizal plants have stronge resistance	high cost and high technology, easy to be affected by more factors

EXISTING PROBLEMS

Physical and chemistry remediation can repair the oil pollution rapidly and efficiently, but have the disadvantages of high cost, secondary pollution and destroying the soil structure, etc. How to apply them into large-scale restoration practice needs further research.

Bioremediation has many benefits, but in actual application it's vulnerable to the natural environmental conditions, leading to unstable and regional repair efficiency. In addition, the microbes can't degrade all the pollutants in soil. The composition complexity of oil pollutants (oil-heavy metal compound pollution), high refractory (the PAHs with high molecular weight, colloid and asphalt etc) and insolubility (such as metal compounds, etc.) greatly enhanced the difficulties of petroleum remediation.

CONCLUSIONS AND PROSPECTS

In a word, the restoration of petroleum-polluted soil is a complex project. At present, the physical and chemical repair methods mainly use heat treatment and chemical leaching methods, which can treat large-scale polluted soil quickly. While some other treatment technologies with low-cost and no secondary pollution are also needed to be developed, especially for the quick emergency response, such as well blowout, oil leakage accident etc.

Now bioremediation technology has made certain achievements. Through large numbers of literature research, it was found that the indigenous microorganisms can keep high activity and degradation efficiency, but too few in natural environment that couldn't meet the actual demands, therefore, how to separate, sift and cultivate more efficient in-situ biodegradable microorganisms is a hotspot. As for phytoremediation, the removal of most pollutants is mainly attributed to the degradation of rhizosphere microorganisms or transformation, thus establishing an effective soil-plant rhizosphere microorganism repair system is of great significance.

In reality, the composition of petroleum pollutants is much more complex, most of soil residues are high-molecular weight PAHs and non-hydrocarbon compounds, maybe accompanied by soil salinization and heavy metal pollution. Given to this, single repair method is difficult to solve the problem thoroughly, therefore, looking for a highly efficient and stable microbes-plant-physiochemical joint repair system which is suitable for the study area and how to convert the technology into reparation of non-point source pollution have a quite brilliant application prospects.

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Biography: Shuguang Wang (1967-), male, senior economist, mainly engaged in land reclamation and soil remediation work.

Corresponding author: Yan Xu (1989-), female, postgraduate, mainly engaged in petroleum geology work.

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