

## DISTRIBUTION AND MOBILITY OF PETROLEUM HYDROCARBONS IN SOIL: CASE STUDY OF THE SOUTH PARS GAS COMPLEX, SOUTHERN IRAN\*

A. AKBARI<sup>1</sup>, M. ARDESTANI<sup>1</sup> AND J. SHAYEGAN<sup>2\*\*</sup>

<sup>1</sup>Graduate Faculty of Environment, University of Tehran, I. R. of Iran

<sup>2</sup>Dept. of Chemical and Petroleum Engineering, Sharif University of Technology, I. R. of Iran  
Email: shayegan@sharif.edu

**Abstract**– This article discusses the distribution of total petroleum hydrocarbons (TPH) contamination in soil within areas of the South Pars Gas Complex (S.P.G.C.), located on the northern shore of the Persian Gulf. To assess the potential risk to groundwater, in situ contaminant mobility was also examined in vertical soil profiles. Sampling was conducted in two rounds, and TPH was determined in accordance with standard methods of TPH analysis. The maximum detected concentration was  $10268 \pm 785$  mg/kg in the area of the waste disposal site. Significant heterogeneity in natural attenuation of different hydrocarbon compounds during transport in the soil was observed. Although the groundwater samples did not show evidence of contamination, vertical contamination profiles clearly showed that groundwater contamination will occur in the near future if no remedial action is taken immediately. It is worth noting that the refineries have been brought to production just in the last 5 years. Also, relevant soil and contamination properties were investigated to recommend an appropriate remediation approach. Bioventing and biopiling were suggested as remediation technologies. However, the result of this study highlights the necessity for national regulations to address cleanup standards and the preventive and remedial guidelines for petroleum hydrocarbon contamination in soil or groundwater.

**Keywords**– Contamination, total petroleum hydrocarbon (TPH), soil, gas refinery

### 1. INTRODUCTION

Hydrocarbon contamination has been recognized as one of the most serious environmental threats arising from the exploration, refining and transport of oil and gas resources. In terrestrial environments, the quality of soil, in terms of biological or physico-chemical properties, would be altered as a result of hydrocarbon contamination. Adverse effects of hydrocarbon contaminants on the soil geotechnical properties such as optimum water content or permeability have been widely reported [1-4]. On the other hand, increased microbial activity has been observed as a result of hydrocarbon contamination. In fact, the observed increase in microbial activity comes at the expense of species diversity [1, 5-9]. In addition, soil pollution always poses a serious potential risk to groundwater quality. The most important toxicological and health implications of hydrocarbon contaminants are attributed to their mutagenic, carcinogenic and toxic characteristics. The toxicity properties influence the living organs in the unsaturated zone [10-13].

Increasing industrial oil and gas activities in the South Pars Gas Complex (SPGC) located on the Iranian shore of the Persian Gulf have negatively impacted the environment of this area. Unfortunately, these deteriorating environmental conditions have not been addressed by current governmental policies. Although a few studies have already investigated environmental issues such as solid waste management

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\*Received by the editors November 14, 2011; Accepted July 10, 2012.

\*\*Corresponding author

problems in the area [14-16], little is known about hydrocarbon contamination of soil and groundwater in this region.

In this study we investigated residual Total Petroleum Hydrocarbon (TPH) contamination in the surface and subsurface soil and groundwater of phases 1 to 10 of SPGC. Then, vertical mobility of the contaminants in the soil and the associated risks of groundwater contamination were assessed. Relevant soil and contamination characteristics that are important in the remediation approach were also studied. Either biopiling or bioventing was proposed as remediation technology. The results of the study provide the baseline data that is necessary for a groundwater monitoring program as well as a remedial plan. Moreover, it is expected that these results will not only alert the company about the need to enforce more stringent procedures in terms of safety and environmental management requirements, but also to encourage the government to establish appropriate regulations, standard guidelines and clean up levels.

## 2. MATERIAL AND METHOD

### *a) Site description*

The SPGC area is located on the northern shore of the Persian Gulf, 570 km from Bandar Abbas. The company was established in 1998 for up and downstream processing of the gas stream originating from the South Pars gas field. The South Pars is the Iranian part of the North Field, which is the largest non-associated gas reservoir in the world, containing 6% of the world's total gas resources. The reservoir is located at a depth of 3000 m and is 100 km offshore from Iran in the Persian Gulf [17]. The SPGC has been designed in 24 phases to provide for the needs of petrochemical industries in the vicinity of the complex and also to export gas condensates, LPG and sulphur [18]. The total area of the SPGC is 140 km<sup>2</sup>, but since only phases 1 to 10 have been brought to production up to now, the investigated area in this study, as shown in Figure 1, was restricted to these phases.

A residential area, Nakhle Taghi village, is located between the complex and the sea. The population of the village is 12000; most of the residents are employed by various active industries in the area. The weather in the region is hot and humid most of the year. The annual monthly average temperature in the area is 28 °C, with a high temperature of 48°C in June and low of 9°C in January. The annual average rainfall is 232 mm with a maximum of 67 mm in December and minimum of 0 mm in June. The annual mean relative humidity in the area is about 60%. [19]

### *b) Soil sampling*

In the first round of sampling, 41 surface soil samples were collected from depths of 0-1.2 m. The sampling locations were determined based on interviews with SPGC personnel and previous records of accidents or leakages from process equipment, storage tanks, pipelines or waste disposal sites. To avoid cross contamination, standard soil sampling protocol was employed in the collection of samples.

In the second round, TPH concentration as a function of depth was examined in detail by drilling 15 boreholes with maximum depth of 22 meters. The drilling points were determined based on the results of the first round of samples. A standing hydraulic drill, Craeilus D-750, and a crane were used for drilling and transferring the drill.

Moreover, 40 groundwater samples were taken from the operating and non-operating wells in the southern border of the SPGC area, north of the Nakhle Taghi village. Several background samples were also collected upstream from the refineries.

A simplified map of sampling locations is illustrated in Fig. 1. Prior to analysis, soil samples were stored at -4 °C.

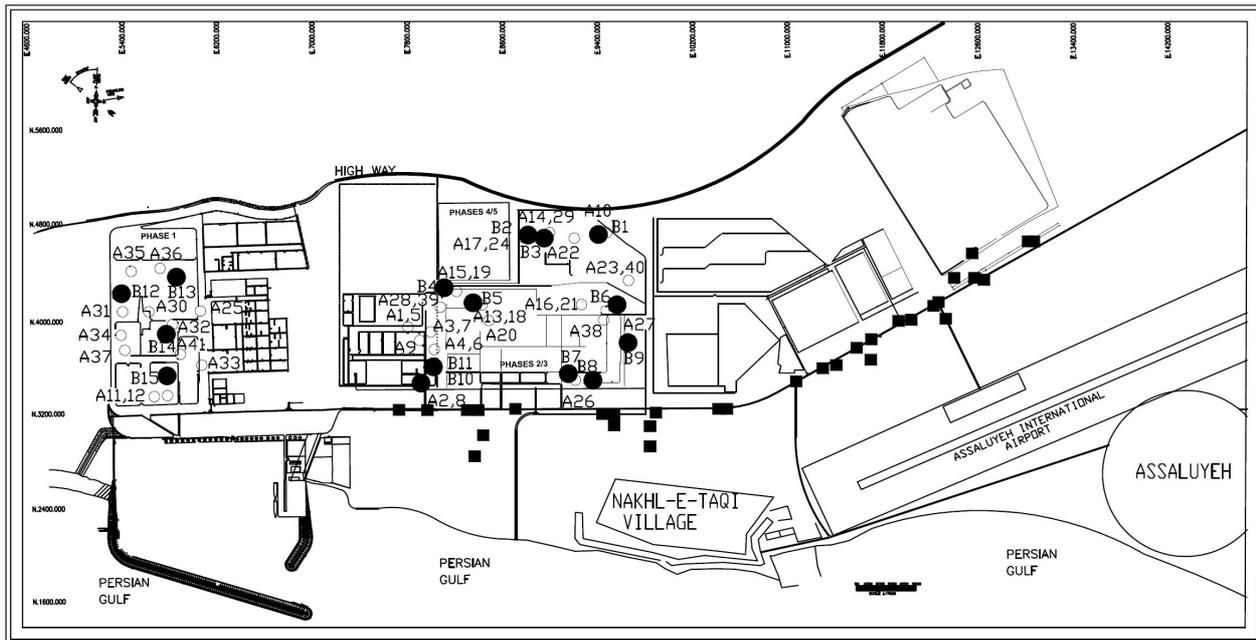


Fig. 1. The locations of sampling points, soil (first round) ○, soil (second round) ●, and groundwater ■

### c) TPH analysis

Soil samples were analyzed in accordance with the USEPA 413.2 analytic technique for total petroleum hydrocarbon using an InfraCal TOG/TPH Analyzer, Model CVH. Methylene chloride was used as extraction solvent and sample clean up was performed by a silica gel-sodium sulphate column to remove the natural organic matter and water from the extraction solution. As this method is more appropriate for screening purposes, extracts of samples with higher contamination levels were analyzed by GC-MS. The GC-MS analyses were performed using an Agilent Technologies 6890N-GC coupled with an Agilent -Mass selective detector 5973N mass spectrometer with an HP-5ms, 30m, I.D=0.25 mm, film 0.25 column. The oven temperature program was 40 to 300 °C at 10 °C/min, followed by an isothermal stage at 300 °C for 10 minutes. The carrier gas was helium with a flow rate of 1 mL/min. The MS was run in selected ion monitoring (SIM) mode using  $m/z$  57 with a source temperature of 230 °C and quadrupole temperature of 150 °C.

### d) Water TOC analysis

The TOC analysis of groundwater samples was carried out using a Skalar TOC analyser (Formacs HT model) on the same day of sampling. This device consists of two channels, one for total carbon (TC), and the other for inorganic carbon (IC) combustion. Carbon dioxide produced from TC and IC combustion was detected by a Non-Dispersive Infrared Detector. (NDIR)

### e) Soil analysis

A soil permeability test was carried out based on the ASTM D-2434 method for 15 samples, 1 sample from depths less than 5 m in each one of the 15 boreholes. Standard particle size analysis was also performed based on the ASTM D-422 method.

Well logs of 15 boring holes showed a relatively similar pattern in all wells as light brown, dense sandy gravel and some silt for depths of 0-12 m, dense sandy gravel and some silt and cobbles for depths of 12-22 m, and finally sand for depths of more than 22 m.

The pH of the soil was measured in suspension in distilled water (1:3) using pH meter Schott CG 825. To determine the soil respiration rate of the soil samples, 10 g of the samples were placed in 20 ml sealed

vials and incubated at 25 °C. The samples from head space were analysed for CO<sub>2</sub> concentrations using a gas chromatograph equipped with a TCD column. Respiration rates were reported in mg CO<sub>2</sub>/kg soil/h, the details of the analysis procedure can be found in Paton et al., 2006 [20]. Soil microbial counts of viable and culturable microorganisms were estimated with the plate count method based on the procedure described elsewhere [21]. Briefly, suspensions of soil and 0.28% sodium pyrophosphate were prepared. Different dilutions from the sample taken from the supernatant were spread onto agar plates. To enumerate total heterotrophic microorganisms, R2A agar plates were used. Purified agar plates supplemented with yeast extract and 20 µL of diesel oil as carbon source were used to estimate the population of hydrocarbon degrader bacteria. Microbial populations were enumerated after two weeks of incubation at 25°C.

### 3. RESULTS AND DISCUSSION

#### a) TPH distribution

Surface soil samples from the sites shown in Fig. 1 were analyzed for TPH. The results are listed in Table 1. Elevated levels of TPH were evident in all of the samples taken from the liquid waste disposal site, SA14, SA29, SA17, and SA24 in the northern area of phase 1. The range of TPH concentrations was 288-5015 mg/kg. Frequent accidents or fires that occur during the transport or storage of waste liquid containers were the main sources of contamination in this area, resulting in a non-uniform contamination distribution with localized highly concentrated spots. These spots pose a serious threat to the groundwater. The ratio of n-octadecane to phytane, and the percentage of n-alkanes (with an even number of carbons) from TPH of sample SA29 are 2.3 and 8.2%, respectively. These values indicate relatively fresh contamination. The age of contamination is important because it is generally known that weathering of contamination decreases the chance for successful and rapid remediation of contaminated soil [22].

Table 1. TPH results of the samples collected in the first round

Sample ID	Depth (m)	TPH <sup>(a)</sup> (mg/kg)	Sample ID	Depth (m)	TPH (mg/kg)	Sample ID	Depth (m)	TPH (mg/kg)
SA1	1.2	43 ±4.1	SA15	1	39±4.5	SA29	0.5	5015.3±421.2
SA2	1.2	N.D. <sup>(b)</sup>	SA16	0.5	84±9.6	SA30	0.8	52±7.3
SA3	1.2	46±4.9	SA17	0.5	288±36.7	SA31	1	52±6.2
SA4	1.1	50±5.5	SA18	1	60±9.6	SA32	1.1	37±2.8
SA5	0.1	<i>1857.6±230.3<sup>(c)</sup></i>	SA19	0.4	176±20.2	SA33	1	N.D.
SA6	0.8	<i>5410.8±410</i>	SA20	1.2	56±7.8	SA34	1	N.D.
SA7	0.7	N.D.	SA21	1	101±12.2	SA35	1	5±1.1
SA8	0.8	N.D.	SA22	1.1	10268.9±785.4	SA36	0.5	64±5.7
SA9	1.2	N.D.	SA23	1.1	N.D.	SA37	1	N.D.
SA10	1.1	N.D.	SA24	1.2	47±5.3	SA38	1.2	N.D.
SA11	1.2	359±12.5	SA25	1	7±1.2	SA39	0	N.D.
SA12	0.5	<i>1270.9±103.7</i>	SA26	1	227±30.2	SA40	0	39±4.6
SA13	0.5	22±3.7	SA27	1	49±3.2	SA41	1	N.D.
SA14	1.2	103±8.2	SA28	1.2	62±5.7	SA42	0	299±42.3

<sup>(a)</sup> TPH values are the average of 5 replicates.

<sup>(b)</sup> Not detected.

<sup>(c)</sup> Italic numbers represent TPH values determined by GC-MS.

TPH contamination was also observed in samples taken from the stabilized (neutralized) solid waste disposal site, located in the vicinity of the liquid waste storage site. There were 7 disposal pits with the dimensions of 3×6×3 m×m×m that were susceptible to leakage. TPH concentrations of the samples taken from this area were in the range of 151- 10200 mg/kg. Evidence of highly weathered contamination was observed in the chromatograms of the samples, as almost 97% of TPH consisted of UCM (unresolved complex mixtures). Also, the detected hydrocarbons were in the range of C14-C28, revealing that volatile fractions had already disappeared.

Moreover, hydrocarbon contamination was detected in the samples taken from the stabilization ponds area, SA13, SA18, SA20, which is located in the central part of phases 2 and 3. Three ponds with the dimension of 3×3×5, m×m×m were used for sludge stabilization by adding lime. It is worth mentioning that no geo membrane was installed to capture the leachate in the disposal site or the stabilization ponds.

A fire training site is located just beside the stabilization ponds. Diesel or gasoline spills occurring during fire simulations have always been a common source of TPH contamination at these training sites [23-25]. As illustrated by Table 1, TPH level of the samples taken from this area, SA15, SA19, SA42 were in the order of several hundred.

Another highly contaminated sample, SA5, was collected from the area around the furnaces at phase 1. The contamination originates from improper furnace fuel handling and storage. However, SA1, the sample taken from depth of 1.2 m at the same point, showed significantly lower levels of contamination, indicating a lower extent of contamination in this area. Not surprisingly, the fraction of higher molecular weight of hydrocarbons, >C18, was dominant in these samples, consisting of 57% of TPH. Moreover, the ratio of heptadecane to phytane was about 0.3, indicating moderately weathered contamination.

High concentration of TPH was also detected in the sample taken from area 2 in phase 1, sample SA6. The contamination was the result of breakage of underground pipes. Again, the extent of contamination was restricted, as in sample SA4, taken 10 m away from SA6, the contamination level significantly decreased to just 5% of SA6's concentration. The ratio of normal heptadecane to phytane for the samples taken from this spot was 2.7, which indicates relatively recent contamination occurred at this point. Moreover, 83% of the total hydrocarbon content of the soil samples were the compounds having boiling points less than the boiling point of n-C18. This implies that there is high potential for the successful application of volatilization- based remediation technologies.

Finally, of the remaining sampling points examined, significant TPH concentrations were evident in samples SA26, SA11 and SA12. The TPH concentration of SA26, taken from the southern area of phases 2 and 3, was 227 mg/kg. Leakage of an underground waste line was the main reason for contamination in this area. The other samples, SA11 and SA12, were taken from the southern part of phases 4 and 5, close to the API unit of the wastewater treatment plant. Highly elevated TPH level of these samples was the result of leakage from the unit, which had been ongoing for two years before finally being fixed.

Considering the sour nature of the gas in the South Pars field, it was not surprising that a strong sulphur odour could be smelled from some of the samples such as SA26, SA11, and SA12.

### ***b) Contaminant mobility***

No TPH contamination was detected at any depth from 0-11 m in 7 of the profiles examined as SB6, SB8, SB10, SB11, SB12, SB13, and SB14. On the other hand, evidence of TPH migration was observed even for low level contaminated profiles.

Significantly elevated TPH concentration was observed in the samples collected from the SB7 profile. SB7 was located in the southern part of phases 2 and 3. The source of contamination was the leakage of an underground waste line at the depth of 6 m. TPH concentration as a function of depth is shown in Fig. 2. As illustrated in this figure, the contamination reached the depth of 20 meters, which was just 250 cm

above the groundwater level. Consequently, groundwater contamination will most likely occur in the near future. However, the water sampled at this point did not show any contamination higher than the detection limit. As is shown in the graph, the concentrations of normal alkanes in the range of C14-C28 were significantly decreased at the depths of 9-11 m. On the other hand, the TPH and phytane profiles had a similar trend of decreasing from the top to bottom of the soil layer. It is generally known that n-alkanes are the first group of compounds that are attacked by microorganisms. Conversely, isoprenoid compounds are more resistant to biodegradation. Consequently, the ratio of octadecane to phytane is usually considered an indicator of biodegradation for hydrocarbon contamination [26]. The ratio of normal octadecane to phytane was 0.8, 0.59, 0.07, 0.07, 0.96, and 0.29 at 6, 9, 11, 14, and 18 m depths, respectively. This could contribute to the heterogeneous biological activity in the subsurface and the higher activity at depths of 9-11 meters.

The ratio of lighter hydrocarbon fractions < C18 to total hydrocarbon content of all of the samples taken from SB7 at different depths was about 50%, suggesting that a combination of volatilization and biodegradation would be effective as a remediation approach. Moreover, the constant ratio of semi volatile fraction to non-volatile fraction at different depths implies that the role of volatilization has been insignificant in the natural attenuation of hydrocarbons, or at least the extent of volatilization has been similar at different depths.

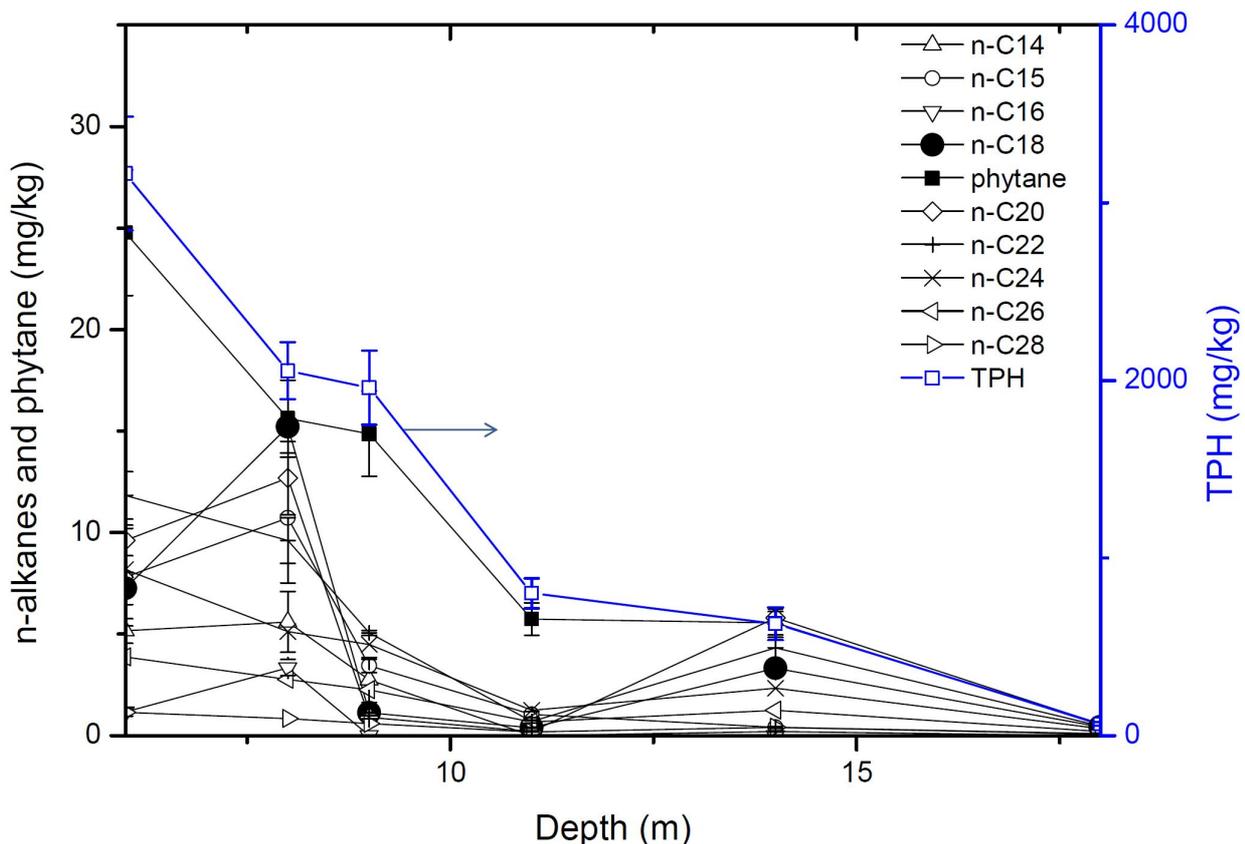


Fig. 2. Concentration of different hydrocarbon compounds and TPH at different depths. Error bars represent the standard deviation of three samples

Three boreholes, SB1, SB2, SB3, were drilled in the northern area of phases 2 and 3. SB1 was located at the solid waste disposal site and SB2 and SB3 were drilled in the liquid waste storage site. As is shown in Fig. 3a, the detected penetration depth of contamination was about 12 meters for SB1. While SB3 had a similar TPH profile with SB1, both the TPH concentration and penetration depth were lower for SB2. The

difference between SB2 and SB3 was not surprising, considering the non-uniform nature of contamination distribution at the liquid waste storage site.

Another borehole, SB4, was drilled in the fire training site, where the TPH concentration decreased from 160 mg/kg at the surface to 40 mg/kg at the depth of 6 meters. The SB5 borehole was drilled in the sludge stabilization area. The TPH concentration profile by depth for this borehole is shown in Fig. 3b. As illustrated in the figure, there was an unexpected contaminant peak at the depth of 11 meters in the SB5 profile. This was because of the change in the soil texture and lower permeability of the soil beyond this depth (permeability of  $1.15 \times 10^{-4}$ ), resulting in the accumulation of contaminants and also a reduced penetration depth. The low permeable soil acted as a barrier protecting groundwater, the level of which was about 30 meters. As the soil texture above the groundwater table in most of the other boreholes was similar to the soil of SB5 from depths of 1-10 m, it could be concluded that in most parts of the SPGC area, the soil was conducive to contamination. This is also consistent with the permeability test results, which indicated that the average permeability of the soil samples in depth 1-8 m was  $4.85 \times 10^{-3} \pm 0.0003$  cm/s. This value is categorized as medium to high permeable soil.

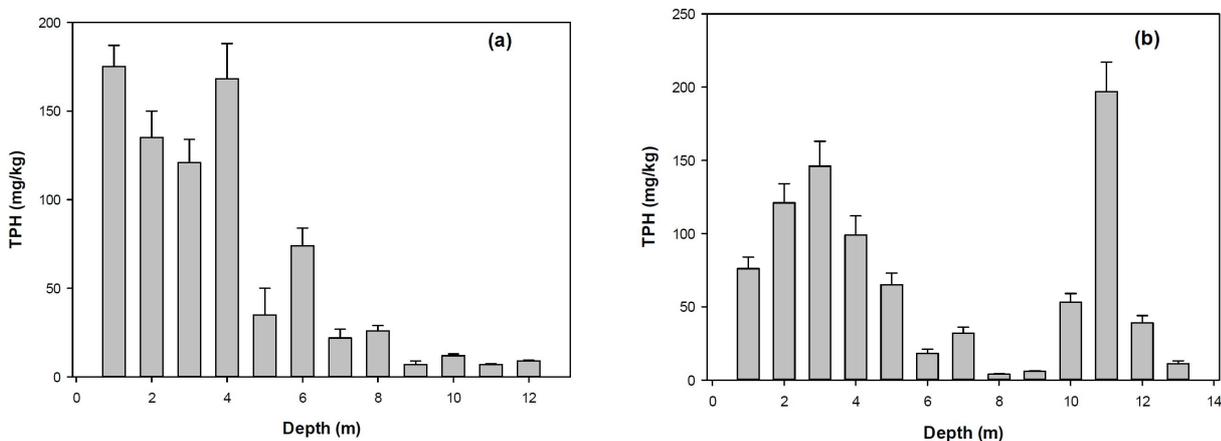


Fig. 3. TPH concentration at different depths of a) SB1 and b) SB5. Error bars represent the standard deviation of three samples

Also, evidence of low level contamination was observed in SB9, which was drilled in the eastern border of Phases 2 and 3. The source of contamination was the leakage from Mono Ethylene Glycol (MEG) pumps. The maximum TPH concentration and penetration depths were 34 mg/kg and 6 meters, respectively. The last borehole, SB15, was drilled around the API unit. Based on the TPH profile of SB15, surface samples were not contaminated, however, contamination was detected in samples taken from the depths of 10-15 m. As the drilling point is 15 m away from the leakage source, this profile clearly shows the extent of horizontal TPH dispersion in the soil.

As discussed, it can be deduced that groundwater contamination is quite possible in the near future. From an environmental health point of view, this situation would be critical as there is a residential area, Nakhle Taghi village, just 750 m downstream of the southern border of the refineries, e.g. the SB7 spot. In the village, groundwater is used for irrigation and drinking after desalination treatment, which is obviously not sufficient to remove hydrocarbon contamination. Although the statistical analysis of TOC results of the groundwater samples from upstream and downstream of refineries did not show any evidence of groundwater contamination at this time, the necessity of applying a monitoring program to assess the water supply is obvious. Moreover, a remedial action program should be planned to immediately stop pollution leaks and then to remediate soil contaminations. Installing geo membranes in stabilization ponds

or disposal sites using gas as an alternative fuel in fire fighter training and also cyclic inspections could be the primary steps of a remedial plan. Unfortunately, currently there is no documented standard for a cleanup level or site assessment of hydrocarbon contaminated sites in Iran. The result of this study highlights the need for more stringent safety and environmental protocols at the SPGC and also the need for the Department of Environment to create national regulations. These regulations should address both the preventive and the remedial standard guidelines for petroleum hydrocarbon contamination of soil or groundwater.

### c) Remediation proposal

To assess the feasibility of the biological approaches for remediation of the contaminated soil, some of the relevant soil properties were evaluated. The soil respiration rate in terms of CO<sub>2</sub> production rate as an indicator of intrinsic biological activity of the soil was determined as 0.056 mg CO<sub>2</sub>/kg soil.h in the first 24 hours for an uncontaminated soil sample, indicating significant biological activities of intrinsic soil biota. Moreover, the microbial population for heterotrophs and hydrocarbon degrader bacteria for representative contaminated and uncontaminated soil samples were determined. The results of plate count experiments and some other characteristics of the site are summarized in Table 2. Comparing this data with the recommended ranges to achieve effective bioremediation[27] indicates the potential for successful bioventing at the site. Moisture and nutrients could be added to stimulate indigenous microbial activities (biostimulation) [28]. For the sites with a higher risk of groundwater contamination, which necessitates immediate soil excavation such as the southern parts of phases 2 &3, the SB7 site, biopiling is proposed to immediately remove the source of groundwater contamination. On the other hand, bioventing could be applied for the other contaminated spots. Bioventing and biopiling both rely on simultaneous volatilization and biodegradation as removal mechanisms for hydrocarbons [29, 30]. The estimated volumes of contaminated soil at different sites are summarized in Table 3.

Table 2. Properties of the soil

	pH	Microbial Presence		Intrinsic permeability cm <sup>2</sup>	Moisture	Maximum TPH level (ppm)
		Total heterotrophic bacteria (CFU/g) <sup>(a)</sup>	Hydrocarbon degrader bacteria (CFU/g) <sup>(a)</sup>			
Contaminated soil		$1.2 \pm 0.88 \times 10^6$	$4.74 \pm 2.6 \times 10^5$		2-10%	<10000
Uncontaminated soil	7.05	$6.12 \pm 0.34 \times 10^3$	$1.47 \pm 0.18 \times 10^3$	$>10^{-8}$		
Recommended range for bio venting	6-8	$>10^3$		$>10^{-8}$	40-60 % of water holding capacity	< 25000

<sup>(a)</sup> The plate count results are the average of 3 samples.

Table 3. Estimated volume of contaminated soil

Site		Estimated volume (m <sup>3</sup> )	Maximum level of contamination (mg/kg)
Solid waste disposal site (SA22)	area source	1800	10200
Liquid waste storage area (SA29)	area source	2000	5000
Phase 1, area 5, close to furnaces (SA5)	area source	70	1800
Phase 1, area 2 (SA6)	point source	500	5500
API unit, phases 4 & 5 (SA12)	area source	400	1200
Southern of phases 2 & 3 (SB7)	point source	1000	3200

#### 4. CONCLUSION

1. The results of the site investigations showed only hydrocarbon contamination at 5 different sites in the SPGC area, with TPH values in the range of 2000-10000 mg/kg.
2. Significant heterogeneity in natural attenuation of different hydrocarbon compounds during transport in the soil was observed.
3. Based on vertical TPH profiles, groundwater contamination will occur in the near future if no remedial action is taken immediately. Contamination was not detected in groundwater samples at the time of sampling. However, TPH profiles indicated the soil's high conductivity to hydrocarbon contaminations in the SPGC area.
4. As hydrocarbon contamination of soil and groundwater is not currently addressed in the national environmental regulations of Iran, the results of the present study highlights the need to create national regulations and standards for preventive and remediation programs by the Department of Environment.
5. Based on the relevant characteristics of soil and contamination, bioremediation technologies such as biopiling and bioventing were proposed as a remediation approach for contaminated soil.

**Acknowledgements:** The authors would like to express their gratitude to South Pars Gas Co. for their moral and financial support of this project.

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