

## **Restoration of Oil Contaminated Soil by Bioremediation for Ground Water Management and Environment Protection**

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### **ABSTRACT**

Water is most essential but scarce resource in our country. Total water resources include surface water and ground water. Groundwater which is the main source for irrigation and drinking in the most part of the world constitutes about 89% of the total fresh water resources in the planet. But in recent years, due to over exploitation of ground water and erratic nature of monsoon, there has been depletion of ground water across the world. Further the quality problems of drinking water – both due to geogenic factors leading to chemical contamination like excess fluoride, arsenic, iron, salinity, nitrate, etc. and anthropogenic factors resulting in bacteriological contamination, pose serious public health problems.

The problem has been further aggravated by the Petroleum industries which generate huge amount of wastewater, solid waste and sludge, some of which may be considered hazardous because of the presence of toxic organics and heavy metals. Accidental discharges as a result of abnormal operations or leakages due to flow line rupture or effluent water carry over with storm water can be major environmental hazard. The article discusses the acute water shortage in the North Gujarat categorically in Mehsana where water pollution is increasing day by day due to accidental discharge of effluent water or crude oil during transportation through pipe lines and enters into lakes, streams, rivers, oceans, and other water bodies. They get dissolved or lie suspended in water or get deposited on the bed. This results in the pollution of water whereby the quality of the water deteriorates, affecting aquatic ecosystems. Pollutants can also seep down and affect the groundwater deposits and ground water recharge system. Against this context the present article attempts to analyze the need for sustainable ground water management in India and highlights the attempt made to restore oil contaminated soil by advance technology i.e. technique of bioremediation of crude oil/oily sludge using specialised bacteria. These specialised bacteria have the capability to breakdown and digest crude oil/oily sludge and convert them into harmless products like carbon dioxide, water and metabolites.

This article discusses the development of microbial consortium and a case study of implementation of bioremediation and result obtained through it at Oil and Natural Gas Corporation, Mehsana (Gujarat).

**Key words:** Total petroleum hydrocarbons (TPH), atomic absorption spectrometer (AAS), Group Gathering Station (GGS), central tank farm (CTF), Gujarat Ecology Commission (GEC).

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### **INTRODUCTION**

The petroleum industry is involved in the global processes of exploration, extraction, refining, transporting (often with oil tankers and pipelines), and marketing petroleum products. The largest volume products of the industry are fuel oil and gasoline (petrol). Petroleum is also the raw material for many chemical products, including pharmaceuticals, solvents, fertilizers, pesticides, and plastics. The industry is usually divided into three major components: upstream, midstream and downstream. Midstream operations are usually included in the downstream category.

Petroleum is vital to many industries, and is of importance to the maintenance of industrialized civilization itself, and thus is critical concern to many nations. Oil accounts for a large percentage of the world's energy consumption, ranging from a low of 32% for Europe and Asia, up to a high of 53% for the Middle East. Other geographic regions' consumption patterns are as follows: South and Central America (44%), Africa (41%), and North America (40%). The world at large consumes 30 billion barrels (4.8 km<sup>3</sup>) of oil per year, and the top oil consumers largely consist of developed nations. While producing oil these industries also generate wastewater, oil sludge, drill cuttings, chemical and chemical mud / drilling mud which in turns contribute to water pollution[9].

Industrial wastewater usually contains specific and readily identifiable chemical compounds. During the last fifty years, the numbers of industries in India have grown rapidly. But water pollution is concentrated within a few subsectors, mainly in the form of toxic wastes and organic pollutants. When toxic substances produced from

industries and enter lakes, streams, rivers, oceans, and other water bodies, they get dissolved or lie suspended in water or get deposited on the bed. The effects of wastewater discharged accidentally by these industries are not only devastating to people but also to animals, fish, and birds and affecting aquatic ecosystems. Pollutants can also seep down and affect the groundwater deposits and it also form layer of suspended oil and grease at the top of the land, creates hindrance in ground water recharge system. The facilities to treat wastewater are not adequate in most of the industries in India. Presently, only about 10% of the waste water generated is treated; the rest is discharged as it is into our water bodies. Due to this, pollutants enter groundwater, rivers, and other water bodies[17].

To overcome the effect of water pollutants discharged from oil industries and to restore affected land, contaminated due to accidental discharge of effluent water or discharge of crude oil in transit from wells to GGS/CTF, the Mehsana Area of North Gujarat was considered.

#### Brief about area of Study:

Mehsana district is one of the districts of north Gujarat agro-climatic zone. The district is located on  $20^{\circ}.02''$  to  $24^{\circ}.42''$  north latitude and  $68^{\circ}.08''$  to  $74^{\circ}.48''$  east longitude with an elevation of 92.96 meter above sea level. This district is surrounded by the Banaskantha district in north, Ahmedabad district in south, Sabarkantha district in east and Patan district in west. Mehsana is district headquarter. The Oil and Natural Gas Corporation, a pioneer in oil industry, started its exploration activities in and around Mehsana city of Gujarat in the year 1964. The important commercially producing fields of the Asset are North Kadi, Santhal, Sobhasan, Balol, Jotana, Lanwa, Bechraji, Nandasan and Linch. Starting with meager production 26 TPD during 1968-69, this Asset is currently producing 6600 TPD Approximately[10].

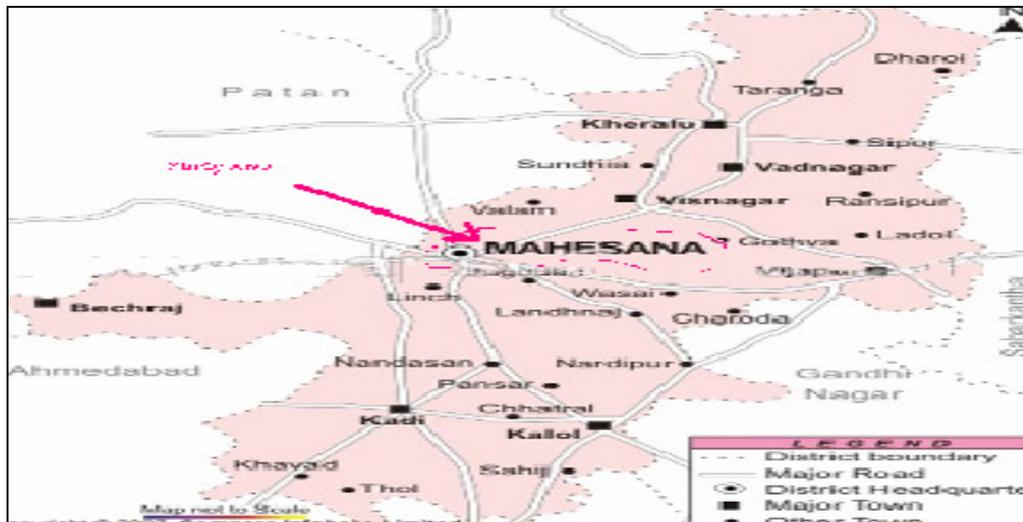


Fig.-1: Site Map

#### Ground water resources in North Gujarat:

As per recent estimate by GEC, 2004 the State of Gujarat has total replenishable ground water resources of 15,811 MCM / yr, whereas utilizable ground water resources for irrigation are 15,020 MCM / yr. The draft from ground water structures is estimated to be 11,486 MCM / yr. The Level of development of ground water resources at present is 76.47%. This leaves a balance of 3,051 MCM / yr for future development of ground water resources[6].

#### Categorization of areas:

The estimation of ground water resources has been carried out considering talukas as assessment units. Based on the level of ground water development the assessment units have been categorized as safe, semi critical, critical and over exploited. Out of 223 talukas in the state of Gujarat 31 units are over exploited (level of GW development > 100%), 12 are Critical (level of GW development between 90 & 100%) and 42 are Semi Critical (level of GW development between 75 & 90%). Over exploited talukas are mostly located in north Gujarat Alluvial plain area. The region wise status of talukas falling indifferent categories is shown in Table No. 1[6].

**Ground water scarcity areas:**

Ground water scarcity areas are mostly located in North Gujarat, Saurashtra and Kachchh regions of the state. In north Gujarat the ground water scarcity areas cover parts of Panchmahals, Banaskantha, **Mehsana**, Gandhinagar and Ahmedabad districts of Gujarat. The scarcity in these areas is faced on account of erratic and scanty rainfall, high level of irrigation development and partly due to inherently saline formations[3].

Beside this existing water scarcity, oily effluent generated by oil industry, spilled over on land, due to accidental leakage from pipe line during transportation.

**Waste disposal and treatment options:**

Many incident of ground water contamination from oil and gas producing industries have been seen across the world. ONGC, Mehsana, which is the largest on shore oil producing asset of Oil and Natural Gas Corporation Limited, India has also observed few such cases and have taken corrective measures to arrest them. Even though if soil becomes contaminated with spilled over hydrocarbons and other waste materials, it is to be disposed off or to be treated by applying various existing methods. The main disposal method is through burial, either on or off site. Treatment occurs via bioremediation (using microorganisms to convert toxic compounds into less toxic forms); or thermal technologies (using high temperatures to reclaim or destroy hydrocarbon-contaminated material). The following types of waste disposal and treatment sites are utilized for oil and gas wastes[9]-

- 1 Pits
- 2 Landfills
3. Land forms
- 4 Land Spreading
5. Thermal Technology
6. Underground Slurry Injection
7. Salt Caverns

**Soil degradation and Bioremediation of oil Contaminated land:**

Soil degradation is widespread in India, affecting, about 1880 lakh hectares or 57% of the total area of the country. Of this 1620 lakh hectares are affected by soil erosion and 250 lakh hectares are affected by in situ degradation (water logging, salinization, and nutrient depletion). Extending the limited data available indicates that area affected by soil degradation in 1947 was probably about 1100 lakh hectares[14].

It is estimated that the cumulative effects of degradation over time will lead to an economic loss of Rs. 8900 crore to Rs. 23200 crore in 1997, reflecting a loss of 11%-26% of annual agricultural output. Erosion has rendered 1180 lakh hectares or 36% of the total area of the country "uneconomical to cultivate because of a significant loss in productivity", while a further 150 lakh hectares has been rendered unmanageable and uneconomical" to use. Erosion by water is the most significant contributor to the economic loss due to soil degradation, and accounts for about 87% of the total degraded area. This category alone accounts for around Rs. 6100 crore-Rs. 21600 crore of the total estimated losses. Salinity and water logging are the other major-kinds of degradation. Although salinization and water logging affect only 11 % of the total area affected by degradation, they account for up to 30% of the total estimated cost. The problems of water logging and salinity are altogether lead to an estimated loss of Rs. 1200 crore to Rs. 2700 crore annually. India, USA, Germany, France, Italy, UK, Japan, China, Russia and South Korea together constitute 68.6% of total oil consumed in the world. India is the sixth greatest energy consumer in the world and Gujarat ranks 1<sup>st</sup>. in on shore oil production 55.10% and 32.3% in gas production in India. The State has highest on shore and offshore oil and gas field (31.3%)[15].

India's crude output in the year to March 2010 is likely to rise 11 percent to 36.71 million tons, or about 734,000 barrels per day [12].

One of the Major contributors of oil and Natural Gas to the country is Oil and Natural Gas Corporation Limited, which is involved in exploration and production of crude oil and Natural Gas, is located in Gujarat.

Oil and Natural Gas Corporation Limited, Mehsana is producing Heavy oil. Heavy crude oil or Extra Heavy Crude oil is any type of crude oil which does not flow easily. It is asphaltic and contains asphaltenes and resins. It is "heavy" (dense and viscous) due to the high ratio of aromatics and naphthenes to paraffin (linear alkanes) and high amounts of NSO's (nitrogen, sulfur, oxygen and heavy metals).

Asphalt is a sticky, black and highly viscous liquid or semi-solid that is present in most crude petroleum and in some natural deposits. The primary use of asphalt is in road construction, where it is used as the glue or binder for the aggregate particles. So the effect of leakage of crude oil or effluent water on land is devastating and affecting ground water recharge system as well. While processing it at GGS/ CTF it also unavoidably generates enormous quantity of tank bottom crude oil/oily sludge as well as oil/oily effluent soaked soil and wastewater which constitutes a major challenge for hazardous waste management as well as environment management.

Due to stringent norms by regulatory authority and corporate responsibility of oil industries to protect the environment, new techniques are continuously in demand in India to manage crude oil/oily sludge. However, in developed countries, new technologies are continuously being invented and implemented for treatment of crude

oil/oily sludge. Several technologies, which were considered emerging a few years back, are now well accepted in the field of hazardous waste treatment. One of new technology is bioremediation of crude oil/oily sludge using specialised bacteria. These specialised bacteria have the capability to breakdown and digest crude oil/oily sludge and convert them into harmless products like carbon dioxide, water and metabolites.

#### **Selection of site for bioremediation:**

The bioremediation project at ONGC Mehsana was undertaken for a total quantity of 1500 tons of oil contaminated soil at the locations of Santhal – 1, near well no 205 and near Well no. 206. The treatment was carried out in three sites at the said locations. The area of the sites is as follows:

- Site – I : Santhal- 1 scrap yard, South Santhal CTF :  
Approximate area = (87 m x 55 m) + (58 m x 20 m) = 5945 m<sup>2</sup>
- Site – II : Near Well no. 205 , South Santhal CTF :  
Approximate area = (40 m x 20 m) = 800 m<sup>2</sup>
- Site – III : Near Well no. 206 , South Santhal CTF :  
Approximate area = (40 m x 22 m) = 880 m<sup>2</sup>
- Total area of the bioremediation site at South Santhal CTF = 7625 m<sup>2</sup> approximately.

## **MATERIALS AND METHODS**

#### **Site preparation for bioremediation job:**

Bioremediation of oil contaminated soil at ONGC, Mehsana, was undertaken to treat total 1500 tons of oil contaminated soil in all the three bioremediation sites. All the three sites were containing heaps of oil contaminated soil. The site preparation was done by spreading the oil contaminated soil on the whole site and leveling the same to a uniform height of 20 cm. The site was cleaned by removing stones, debris, iron scrap material and any other non biodegradables. This was done by using JCBs and tractors with the help of manual laborers. All the sites were properly segregated and identified. The site preparation job was completed by 25 days and then the actual bioremediation job was initiated by the application of Oil zapper on the bioremediation site.

#### **Development & Application of Oil zapper on the bioremediation site:**

Oil zapper is a new bacterial product developed for the purpose of bioremediation of contaminated soil[10]. This was developed by assemble of five bacterial species which could biodegrade aliphatic, aromatic, nitrogen, sulphur, oxygen containing compounds and asphaltene fractions of crude oil/oily sludge. Oil zapper (sludge degrading bacterial culture) was produced in a bioreactor at laboratory. The growth conditions in the bioreactor were as follows: temperature, 32°C; aeration, 0.75 volume of air/volume of medium/min; agitation, 250 rpm; pH 7.0 (adjusted with 1 N HCl-NaOH); and duration of growth, 15 h. Silicone oil was added to control excessive foaming in the bioreactor. After growth, the culture was immobilized onto the selected carrier material, corncob powder, a biodegradable agricultural residue, by simple mixing in a1:3 ratios of carrier material and culture. A total bacterial count (on Luria-Bertani agar [LA] plates) of 10<sup>10</sup> CFU/g of carrier material with a moisture level of 70% was maintained while the bacterial consortium was immobilized. The carrier-based culture was dispensed into sterile reusable polyethylene bags (4 kg of culture immobilized onto carrier material in each 10-kg polyethylene bag) and stored at 4°C after the bag was aseptically sealed. Oil zapper culture (live sludge degrading bacteria) was applied on oil contaminated soil at the Site I, II & III of the bioremediation site. Nutrient mixture was also sprayed on oil contaminated soil. After application of Oil zapper, mixing of oil contaminated soil and Oil zapper bacteria was done by tilling of site using tractor with cultivator.

#### **Sampling at the bioremediation site:**

Total 1500 tonnes of crude oil contaminated soil was undertaken for the bioremediation at site I, II & III of South Santhal .Sampling was done in the sites of the bioremediation site at zero day i.e. before initiation of the bioremediation job and at regular intervals after application of Oil zapper on the bioremediation site till the completion of the bioremediation job. Samples of the oil contaminated soil was collected from the random points of the bioremediation site and collected in a plastic bag. The sampling point was considered by dividing the site in number of zones separated at 20 meter distance and from each zone samples were collected randomly. In Site – I samples were collected from 10 random sampling points whereas in Site – II & III, samples were collected from 5 random sampling points from each sites. Collected samples from bioremediation sites were mixed uniformly with

equal quantity to get a composite homogenised mixture of the samples. These composite samples were analysed for related parameters for monitoring the bioremediation efficiency.

#### **Extraction of TPH:**

The moisture content was determined by heating the oil contaminated soil at 80 °C, where the water layer will be separated from the oil contaminated soil. Total petroleum hydrocarbon (TPH) was extracted from the oil contaminated soil samples by using solvents (hexane, toluene, ethylene chloride and chloroform). Solvents were evaporated in a fume hood by gentle nitrogen stream. After solvent evaporation, the total petroleum hydrocarbon (TPH) in each sludge samples was quantified. TPH extracted from crude oil/crude oil contaminated soil samples were fractionated into alkane fraction, aromatic fraction, NSO fraction and asphaltene fractions by silica gel column. After TPH extraction the residue was further analysed. The residue was taken in crucibles and heated at 600°C in a Muffle furnace for 5-6 hours. After cooling, the amount of ash was quantified [4].

#### **Analysis of selected heavy metals in crude oil contaminated soil:**

Selected heavy metals were analysed in composite samples of crude oil contaminated soil undertaken for the bioremediation collected at zero day and after completion of bioremediation. The sludge sample was digested in nitric acid. A known weight (approximately 1 gm) of sludge sample was taken in a clean and dry container (normally microwave digestion Teflon tube or 100 ml glass beaker). Concentrated nitric acid (approximately 15 ml.) was added to a container and the container was covered with a watch glass and heated at 140°C on a hot plate, in a fume hood, until most of the acid was evaporated. The step was repeated thrice in order to solubilise the metallic components. The solution was then filtered in another container through 0.45 micron Whatman filter paper number 42 and the insoluble residues on the filter paper were rinsed with 10% nitric acid. The residue was then discarded and the container was covered with a watch glass and heated at 140°C until complete evaporation of nitric acid. Now the container was heated to 400°C until barely dried and white ash appeared. The sample was not allowed to bake and the temperature was maintained at 400°C for six hours. The bottom of the watch glass was carefully rinsed into the container using 10% nitric acid. The sides of the container were also rinsed and solution was evaporated to dryness at 140°C. The filter paper blank and nitric acid blank were also prepared similarly. Each container was then cooled and the residue was dissolved in 1 ml of concentrated nitric acid. The clear solution was then quantitatively transferred into 50 ml volumetric flask and volume was made up to 50 ml by using 10% nitric acid. The selected heavy metals present in the extract were analyzed using Atomic Absorption Spectrophotometer (AAS) (AAS – TJA, SOLAAR M Series, Unicam, USA). Some metals such as Se, and as, were analyzed using AAS equipped with hydride generation system or cold vapour technique [10].

## **RESULTS AND DISCUSSION**

At zero days, the oil contaminated soil samples were collected from the bioremediation sites I, II & III of South Santhal CTF, ONGC, Mehsana Asset. The composition of the oil contaminated soil was estimated by the analysis methods mentioned above. As per the analysis it was found that the oil contaminated soil undertaken for bioremediation at the bioremediation site I contained 7.87 % solvent extractable total petroleum hydrocarbon (TPH), 18.51 % moisture / water content, 73.62 % organic heavy fraction of hydrocarbon and inorganic (Table 2). The site II of the bioremediation site contained 7.32 % solvent extractable total petroleum hydrocarbon (TPH), 19.42 % moisture / water content, 73.26 % organic heavy fraction of hydrocarbon and inorganic (Table 2). The site III of the bioremediation site contained 6.92 % solvent extractable total petroleum hydrocarbon (TPH), 18.25 % moisture / water content, 74.83 % organic heavy fraction of hydrocarbon and inorganic (Table 2). It was observed that the steam extractable TPH in the oil contaminated soil in all the three sites were below extraction level (Table 2). Total petroleum hydrocarbon extracted from the oil contaminated soil of the bioremediation site I contained 65% alkane fraction, 24% aromatic fraction, 11% NSO and asphaltene fraction as shown in Table 2. The same in site II contained 64% alkane fraction, 25% aromatic fraction, 11% NSO and asphaltene fraction as shown in Table 2. The same in Site III contained 66% alkane fraction, 24% aromatic fraction, 10% NSO and asphaltene fraction as shown in Table 2. Biodegradation of TPH in oil contaminated soil at the bioremediation sites ONGC Mehsana is given at Table 3 and at Figure - 2

GC chromatogram indicating the biodegradation of Alkane and Aromatic fractions of the TPH extracted from the samples collected from the bioremediation sites of Site – I are also taken and given at Figure – 3 and 4

Reduction in TPH has taken place within a period of 135 days from 7.87% to 0.7% and biodegradation reached to 90.98% at site -I indicate successful in-situ bioremediation of the oil contaminated soil. The land has become cultivated.

The physical result of the bioremediation on the affected oil contaminated sites can also be seen from the photographs taken. (Status Before and after bioremediation are given from Figure -5 to Figure -10).

### CONCLUSION

Bioremediation is a process that uses naturally occurring microorganisms to transform harmful substances to nontoxic compounds. Bioremediation exploits this natural process by promoting the growth of microbes that can effectively degrade specific contaminants. Thus Oil zipper technology utilizes the bioremediation potential of specific microbes that degrades the toxic hydrocarbon compounds leaving behind non hazardous end products or metabolites and hence no harmful effects. Not only this technique is environmental friendly but also highly cost effective when compared to storage of oily waste/oily sludge in sludge pits and removing and transporting oil contaminated soil from the affected site due to accidental leakage of effluent water or crude oil. Further, the technique is ecologically sound, natural process; existing microorganisms can increase in numbers when oily sludge and waste water effluent sludge (the contaminants) is present. When the contaminants are degraded, the microbial population naturally declines. The residues from the biological treatment are usually harmless products (such as carbon dioxide, water and fatty acids) and hence, bioremediation technique could greatly help in solving the problem of soil contamination, oil sludge and waste water management problems of oil industries and controlling environmental and ground water pollution and also providing access to recharge of ground water system to the great extent at places where layer of oil contaminated soil formed restricting water percolation.

Table- 1: Region-wise status of categorization of Talukas. (Source – GEC 2004)

Region	No. of safe Talukas	No. of Sami Critical Talukas	No. Of critical Talukas	No. of Over exploited talukas	Saline Talukas	Total No. of Talukas
North Gujarat	11	14	6	25	7	63
Central Gujarat	33	15	1	1	2	52
South Gujarat	24	1	0	0	0	25
Saurashtra	28	35	4	2	4	73
Kutchh	1	4	1	3	1	10
Total	97	69	12	31	14	223

Table -2: Composition of oil extracted from contaminated soil undertaken for bioremediation at, ONGC, Mehsana

Constituents of oil contaminated soil	Composition (%) in the oil contaminated soil collected from		
	Site – I	Site – II	Site – III
Steam extractable total petroleum hydrocarbon (TPH) in oil contaminated soil	Nil	Nil	Nil
Solvent extractable TPH in oil contaminated soil	7.87	7.32	6.92
Water content in oil contaminated soil	18.51	19.42	18.25
Heavy fraction of hydrocarbons & inorganics	73.76	73.26	74.83
Constituents of TPH			
Alkane fraction	65	64	66
Aromatic fraction	24	25	24
NSO & Asphaltene fraction	11	11	10

Table- 3: Biodegradation of TPH in oil contaminated soil at the bioremediation sites ONGC Mehsana.

Time period	Site I		Site II		Site III	
	TPH (%)	Biodegradation n (%)	TPH (%)	Biodegradation n (%)	TPH (%)	Biodegradation (%)
Zero day	7.87	--	7.32	--	6.92	--
After 15 days	5.12	34.94	5.27	28.01	4.29	38.01
After 40 days	4.21	46.51	3.98	45.63	3.14	54.62
After 75 days	2.04	74.08	2.41	67.08	1.95	71.82
After 135 days	0.71	90.98	0.58	92.08	0.59	91.47

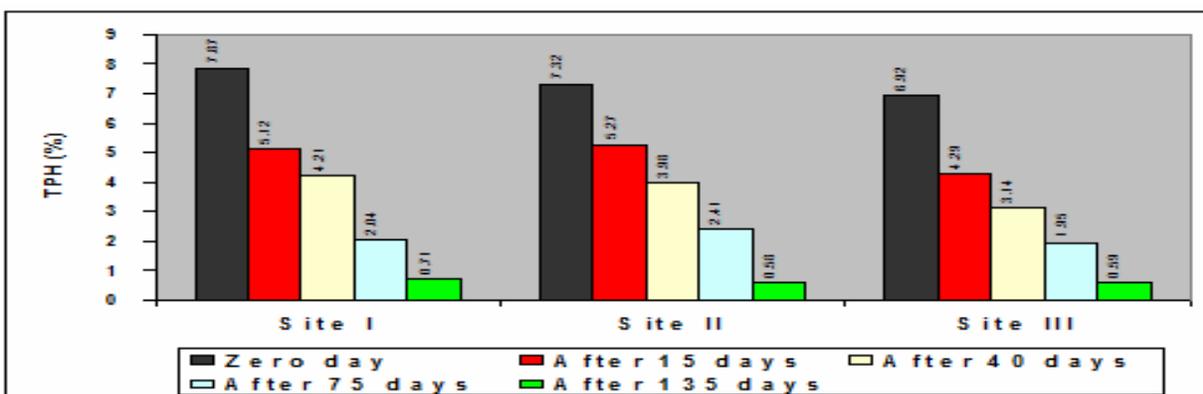


Fig. - 2: Bioremediation of oil contaminated soil at South Santhal CTF, ONGC Mehsana Asset.

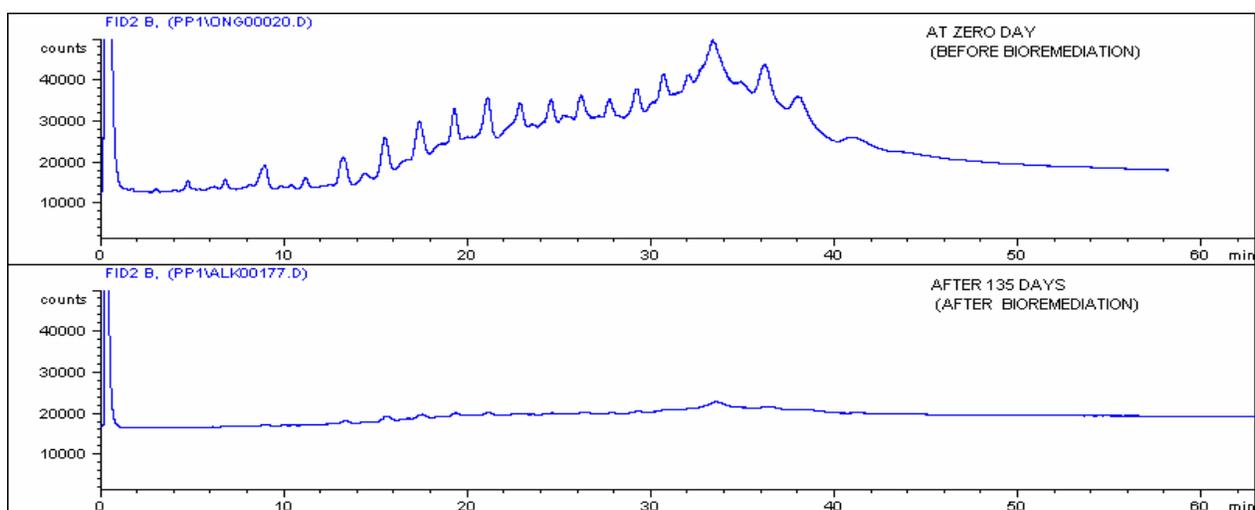


Figure - 3: GC chromatogram indicating the biodegradation of Alkane fraction.

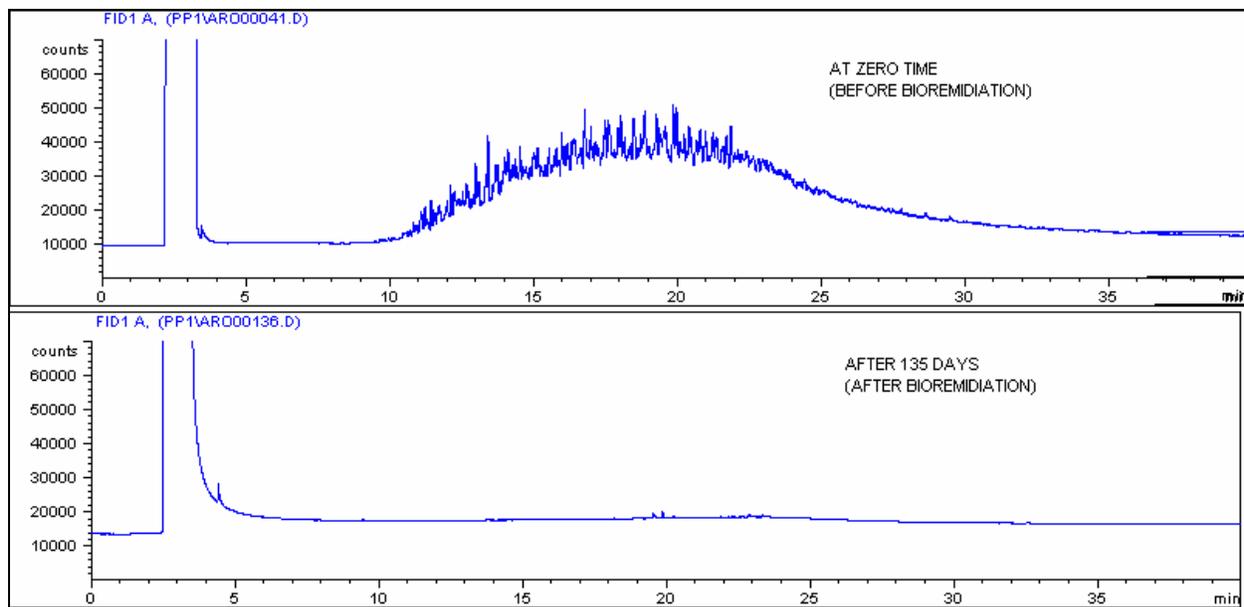


Figure - 4: GC chromatogram indicating the biodegradation of Aromatic fraction.



Fig.-5: Site -I before Bioremediation



Fig.-6: Site - I after Bioremediation



Fig.-7 Site – II before Bioremediation



Fig.-8: Site – II after Bioremediation



Fig.-9: Site – III before Bioremediation



Fig.-10: Site – III after Bioremediation

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