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Bioremediation Techniques for Oil-Sludge Treatment at the Arun Field

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Abstract

lurry sludge that contains hydrocarbons and heavy metals compounds were generated rom drilling activities and production processes in the Arun Field in North Aceh. The ludge was categorized as a hazardous and toxic waste, and it required special handling nd treatment. As a part of the overall waste management program, the sludge was eated by applying bioremediation technology to reduce the concentration of nvironmental pollutants to levels considered acceptable by the Indonesian Regulatory gencies. The pre-treatment sampling result showed that the Total Petroleum Iydrocarbon (TPH) concentration of the sludge was in the range of 2-4% and heavy ıetal compounds such as lead, zinc and mercury were above the government limits.
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‹TRODUCTION

:l and Gas Industries in Indonesia have significant contribution as a leading sector to the untry growth. These industries have multiplier effect for national economic such as eating job opportunity and generating new economic activities that strengthen local mmunity around concession area. However, Oil and gas production processes done by

Aceh Production Operation - ExxonMobil Oil Indonesia Inc (APO-EMOI) have potential impacts to environment. To mitigate potential environment impact of the generated oil sludge from drilling and production activities at Arun Field, APO-EMOI, since 2004, EMOI has committed to use any application of waste treatment environmentally sounds. Bioremediation techniques have been used to treat oil-sludge waste in the existing mud pit at cluster IV, APO-EMOI.

The project aims to reduce the Total Petroleum Hydrocarbons (TPH) and heavy metals contents in the highly contaminated oily sludge in compliance with regulation of the State Minister of Environment Decree No.128/2003 on Procedure and Technical Requirements on the Biological Treatment of Petroleum Waste and Petroleum-Contaminated Soil.
The project is focused on the combined clean-up technique known as lagoon, biopile, and slurry treatments where bio-augmentation and bio-stimulus were simultaneously conducted. The preparation of microbial consortium as the degrading agents will be discussed further in this paper. The microbial consortium was isolated from the existing contaminated sludge.

STATEMENT OF THEORY AND DEFINITION

Bioremediation is believed to be one of environmental-friendly technology to treat polluted sites in the oil and gas industries for more than 50 years as an efficient way to degraded oil sludge (Bartha, 1986). Bioremediation employs microorganisms capable for degrading the target pollutants under certain conditions. The process can be carried out either by the addition of exogenous microorganisms (bio-augmentation) or by stimulating the indigenous microorganisms with the addition of nutrients (bio-stimulus). The hydrocarbon degrading microorganisms can be isolated from various sources, such as polluted sites, compost, and manures. Eventually, the state-of-the-art approach to remediate contaminated soil was to excavate and dispose to a secure landfill. Facing up the liability concerns, costs, and regulatory constraints, soil/sludge cleanup alternative technique is encouraged. Thus, on site permanent solutions are the preferred method of treatment, especially those that involve the complete destruction of the contaminant using biological (natural) techniques, *i.e.* bioremediation. In order to support the bioremediation operation, monitoring of its performance is conducted with some important procedures highlighted as follows:

Physiochemical Analysis
pH was determined as soil's actual acidity (in distilled water) and potential acidity (in a solution of KCl 1N). TPH and Oil & Grease were extracted according to the methodology described in APHA, 20^{th}, 1998 5520-B-C/Infra Red Spectrophotometry. The temperature, Sulfide (H_2S) and Ammonia (NH_3) measurement used thermometry, spectrophotometer (methylene blue), and Indophenol, respectively. Chemical Oxygen Demand (COD) and total Phenol were determined titrimetrically. Heavy metals contents and Total Characteristic Leaching Procedure (Arsen, Barium, Cadmium, Chromium, Lead, Cooper, Mercury, Selenium and Zinc) were measured using Atomic Absorbance Spectrophotometer (AAS). Other parameters were observed (data not shown) including

the carbon organic which is determined according to the Walkley and Black methods, the nitrogen content in the soil measured using the Kjeldahl steam distillation method and Phosphorous which is measured using the total phosphorous. Since climatic data is the most influence the design of mode treatment, rainfall was also observed.

Enumeration and Identification of Bacteria

The isolation of microorganisms from mud pit and a contaminated area were carried out by dissolving 5 g of sample into 10 ml of 0.85% Potasium Chloride (NaCl) solution. It was then plated on both nutrient agar and PDA. Purified colonies were grown individually on a minimal media, which consisted of 2.2 g of K_2HPO_3, 0.73 g of KH_2PO_4, 1 g of $(NH_4)_2SO_4$, 30 g of NaCl, and 0.2 g of $MgSO_4$ with 2% of diesel fuel added as the sole carbon source. To evaluate the microbial population, the samples collected are placed in contact with a 0.85% NaCl solution and shaken for 30 minutes, to extract the bacteria found in the soil or sludge matrices. The total heterotrophic bacteria are counted according to the surface spread method. The hydrocarbonoclastic bacteria count is done using the most probable number method, with diesel serving as a source of carbon identification. Bergey's Manual of Systematic Bacteriology and Bergey's Manual of Determinative Bacteriology are used for identification through microscopically observation and the physiological test (BIOLOG).

Acute toxicity test (LD_{50}; Lethal Dose)

A composite soil/sludge sample from bio-remediated sludge was mixed prior for the analysis of acute toxicity test. The LD_{50} analysis for solid material was conducted in form of 96-hour acute oral toxicity test (lethal dose 50) (OECD, 2001). Adults mice (*Mus musculus*), at least 2 months old (male and female) from the same culture are used as the test species. *Mus musculus* was purchased from reputable supplier (Bogor Agricultural University). In this protocol, *Mus musculus* mortality is used as end point of the primary test up to 15.000 mg/kg body weights (EPA, 2005). Clinical change (*i.e.* gross necropsy) was also observed.

DESCRIPTION AND APPLLICATION OF EQUIPMENT AND PROCESSES

Project Description

The bioremediation project was conducted at the Arun Field, Aceh Production Operation, ExxonMobil Oil Indonesia, Inc from January 2007 to September 2007. During this period, bioremediation of 6.000 M^3 of waste in a mud pit used back hoe for mixing process, air compressor and blower for the aeration supported by electrical generator, various pumps and trucks were deployed for waste transfers. Production of 120 M^3 of microbial consortia culture (MCC) needs the used of organic (cow manure, compost) and inorganic -fertilizer (urea and NPK). Addition of bulking agents such as palm oil husk, rice husk, grass cutting were also necessary to reduce soil bulk density as well as serving as an additional organic material during the bioremediation process. The number of resources involved in this project was 20 man-powers.

Treatment design

As can be seen in the Figure 1, from mud pit, we distinguish the three phase of waste, *i.e.* water, slurry and solid. Such condition allows us to alternate change the modes of operation depending on the real-time condition of the waste and climatic condition. The water was treated as a lagoon treatment and its functions at the same time for water make-up (watering) for slurry and solid treatment. The muddy waste or even already in form slurry are subject for slurry treatment. Some muddy waste can become dried through evaporation against sunlight energy. The dried solid materials were used for pile construction.

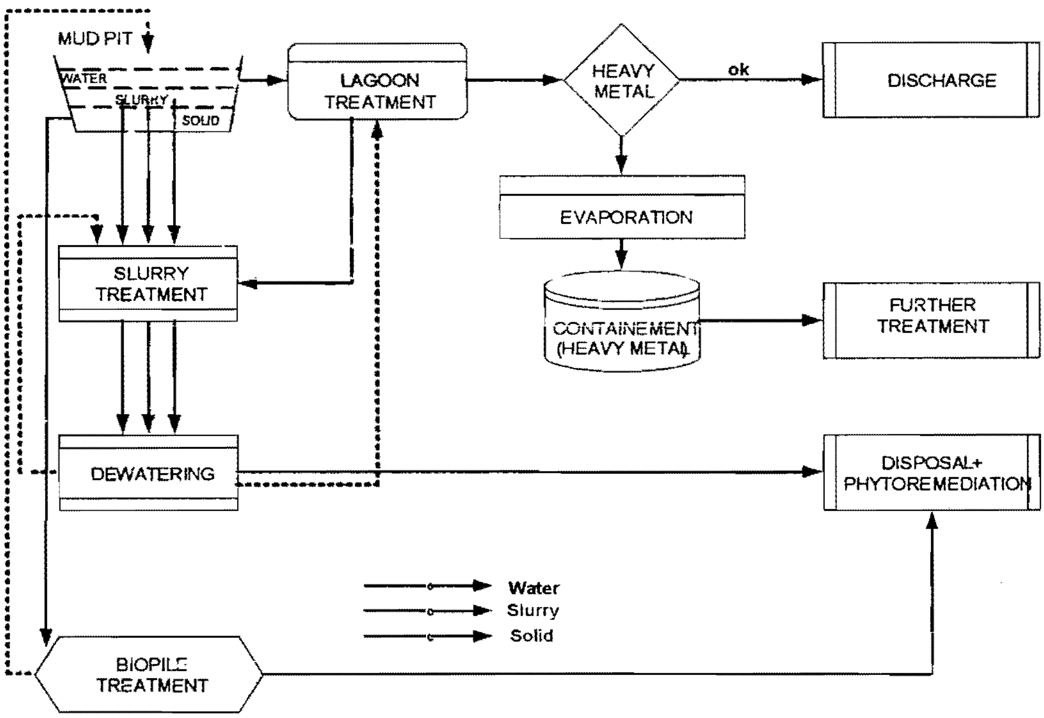

Figure 1. Treatment design of the conducted bioremediation

For all treatment, a quantity of microorganisms in form of microbial consortia (bacteria, fungi, yeast) is necessary. They are ultimately responsible for mineralizing most target pollutants to carbon dioxide. Thus, with the understanding of this naturally occurring process of bioremediation, better contaminant clean-up process objectives could be achieved.

Two metabolic modes namely aerobically and anaerobically of *lagoon*[1], *biopile*[2] *and slurry*[3] treatments were applied for treating the oily sludge from the mud pit. Aerobic method is performed by using air injection to ensure oxygen transfer for organic contaminants degradation process. Indeed, such aerobic bioremediation requires the

[1] involves constructing soil piles above ground, with the contaminated soils placed within the bund area

[2] involves constructing soil piles above ground, with the contaminated soils placed within the bund area

[3] The waste from mud pit was mixed with a predetermined water amount to form the slurry

availability of oxygen supporting the action of enzymes to accelerate the reaction and metabolic pathways of the degradation of petroleum hydrocarbons. Otherwise, the anaerobic method will performed a composting process with the help of additional bulking agents as remedial agents for the contaminated sludge. Moisture, heat, nutrients, oxygen, and pH which plays an important roles in controlling the biodegradation process were maintained in the optimum condition prior to the biological activity of microbial consortia.

PRESENTATION DATA AND RESULTS

Microbial Consortia Preparation

There were more than 17 different microbial colonies isolated from the mud pit. Twelve of them were bacteria such as *Achromobacter* sp, *Acinetobacter* sp, *Aeromonas* sp, *Bacillus* sp, *Echericia coli*, *Flavobacterium* sp, *Koucuria* sp, *Mycobacterium* sp, *Pseudomonas* sp, *Proteus* sp, *Rugeria* sp, and *Sphingomonas* sp. Four were fungi (*Candida* sp, *Cryptococcus* sp, *Rhodotorula* sp, and *Saccharomyces* sp) and one unidentified Actinomycetes. Six of the isolated microbial shown the ability to degrade Poly Aromatic Hydrocarbons (PAHs) which known as persistence and carcinogenic agents. The presence of ***Pseudomonas sp*** is expected since this bacterium is recognized to be able to decompose many organic pollutants. These Gram-negative bacteria are rod-shaped cells with multitrichous flagella. *Pseudomonas sp* is listed among microorganisms most commonly found in various unfavorable environments. Another interesting result is found since ***Sphingomonas*** has been detected in the mud pit samples. Such species is previously described as a marine hydrocarbonoclastic bacteria which is belong to Gram-negative bacteria, which able to grow under aerobic conditions with hydrocarbons as sole carbon and energy source. Whole consortia are probably halo tolerant since most of the test is conducted by using marine gel broth with the salinity around 3000 ppm.

The inoculations of different potential microbial sources were used in order to achieve an optimum biomass with maximum degradation activities. A consideration during the selection of contaminants concentration was based on their maximum possible number of cells. The biodegradation rate expectation was determined and the hypothetical stoichiometric formula was used to estimate required growth factors such as: oxygen, nutrients and pH. Bigger microorganism quantities cultivation added to the treatment as soon as the start up unit reaching desirable targets.

Sludge Treatment

By taking into a consideration that climatic condition and possible stratification in the mud pit (water; slurry and solid), a combination of water lagoon, biopile, and slurry treatments were applied simultaneously.

1. ***Water Lagoon Treatment***

The treatment process of this technique has been done in March, 2007, prior to the water release to the environment. The two cells of lagoon water treatment cells processed

excess water potentially overflowed from mud pit. Blowers were used to maintain aeration and mass water were inoculated by 4 M^3 of microbial consortia culture (MCC).
In most cases, lagoon treatment was enriched by MCC to bloom the number of photosynthetic green algae *i.e. Chlorella* sp. Such green algae, can benefit for advanced treatment sincc *Chlorella* sp was recognized has a great capacity to uptake and accumulate organic contaminants (ammonia, phosphate), heavy metals and even Polychlorinated biphenyls (PCBs). Cyanophyceae blue-green algae *Oscillatoria* sp which has also been identified in the water of lagoon treatment reacted more sensitive to heavy metals than green algae (Kusel-Fetzmann *et al.*, 1989). The number of phytoplactons species were 2.963 cell/mL, 741 cell/mL, 2.774 cell/mL, 185 cell/mL and 16.783.333 cell/mL for *Oscillatoria* sp, *Euglena* sp, non identified flagellated organisms, *Euglena* sp, and *Chlorella* sp, respectively. The test result of some physico-chemical parameters was tabulated in Table 1.

Tabel 1. Test results for environmental release water

Description Test	Water from mud pit	Leachate from mud pit	Standard*	Unit	Method
pH	7.4	7.1	6.0 – 9.0	-	Electrometry
Temperature	28.0	28.0	45	^{0}C	Thermometry
Sulfide, H2S	0.12	<0.01	1.0	mg/L	Spectrophotometry (Methylene Blue)
Ammonia, NH3	0.62	1.62	10	mg/L	Spectrophotometry (Indophenol)
COD	130	240	300	mg/L	Tritimetry
Oil & Grease	< 0.5	< 0.5	-	%	IR Spectrophotometry
Phenol Total	0.27	0.15	2	mg/L	
Heavy metals			**Standard ****		
As	0.05	0.72	5	mg/L	AAS
Ba	7.2	6.9	150	mg/L	AAS
Cd	0.23	0.57	1	mg/L	AAS
Cr	0.75	1.25	5	mg/L	AAS
Pb	0.03	0.46	5	mg/L	AAS
Cu	2.1	2.8	10	mg/L	AAS
Hg	165	185	200	µg/L	AAS
Se	0.35	1.55	1	mg/L	AAS
Zn	1.85	4.75	50	mg/L	AAS

*) Minister Decree: Kep-42/MENLH/10/1996 Annexes 1 (land)
**) Minister Decree: Kep-128/MENLH/2003

The results indicated that oil and grease concentration were less than 0.5 % for both cells. The Chemical Oxygen Demand (COD) of the mud pit samples was below the threshold level, less than 150 mg/l and 240 mg/l respectively for lagoon cell 1 and 2. Since the existing mud pit has a leachate canal (found incidentally at the south side of mud pit), we have to control such leachate by installing overflow sump pit (2 x 1.5. x 0.7) M in dimension.

The finding results were desired in predicted way by the facts that available heavy metals can be absorbed and immobilized in less toxic form within *Chlorella sp* cells. The results indicated high dominance of *Chlorella sp* as a result of *Chlorella sp* biomass was too high in abundance.

2. ***Biopile***

In order to find out the optimum conditions for the biodegradation of oil sludge, contaminated sludge was supplemented with nutrients *i.e* cow manure and inorganic amendments (NPK and Urea), bulking agent (palm husk, saw dusk and cutting grass) and a bacterial consortium as inoculum (MCC). During three months of treatment, about 2000 M^3 of sludge has been treated from three cells of biopile of 200-250 M^3. Figure 2 showed sludge matrix, total petroleum hydrocarbons decreased over the time. The increased oil degradation could be attributed to the selected bacterial consortium comprising of strains of *Acinetobacter sp, Aeromonas sp, Pseudomonas sp, Bacillus sp, Flavobacterium sp, Corynebacterium sp* etc.

The addition of bulking agents when pile has been constructed, tend to have a priming effect on microbial populations. The bulking agent might have played a role in reducing soil bulk density as well as serving as an additional organic material during the bioremediation process. It has also been noted previously that the addition of organic materials to soil enhances oil degradation (Chang and Weaver, 1998). The tillage of soil might have enhanced biodegradation by increasing bioavailability of the hydrocarbons compounds for carbon sources and energy for microorganisms. Small clumps were noted during the early stages of the construction, but disappeared during tillage and this probably enhanced redistribution of the hydrocarbons compounds, making it more available for microbial degradation. In general, the tillage of soil might enhance the contact between sludge containing hydrocarbons and bacterial populations thereby enhancing the bioremediation process.

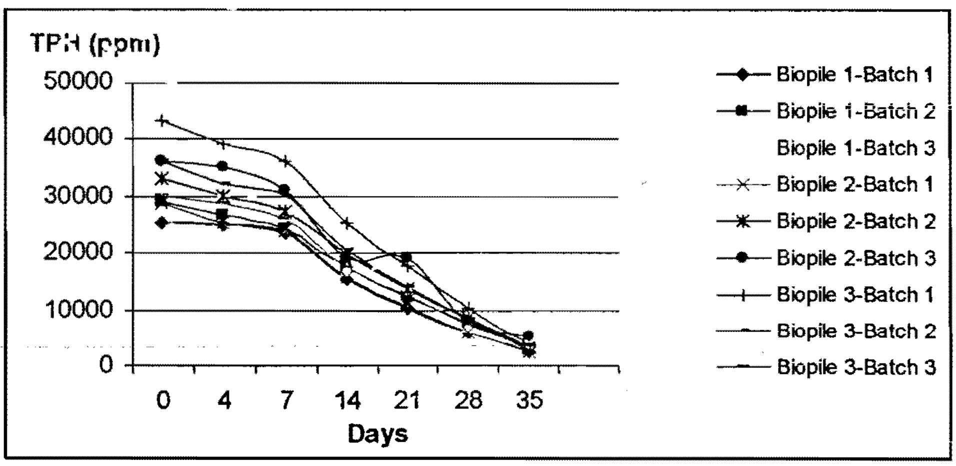

Figure 2. Total petroleum hydrocarbon (TPH) removal in the biopile cells. Indicated value was the average of 3-5 composite samples.

3. Slurry Treatment (mud pit)

Slurry-Phase, directly in the mud pit, was a controlled treatment by mixing sludge with water as an open bioreactor to provide in low viscosity. The waste from mud pit was mixed with a predetermined water amount to form the slurry. The next procedure was the disposition of the mixed slurry (by decantation) for two days prior to the dewatering. Aeration was maintained for maximum periods through blower system. The effectiveness of aeration was optimized by using Wilden pump to transfer slurry material creating aeration mechanically. Other aeration system was done by installing a simple air lift pump through a 1-inch diameter of 12 bio-venting pipes into the mud pit. The system was operated for 4-6 hours per day using air compressor (180 cfm). The maximum rate that may occasionally be resulting the release of contaminants vapor from the pit. After three-four weeks of treatment, bioremediation process results in significant decrease in TPH. The effectiveness of bioremediation shown in the Fig 3.

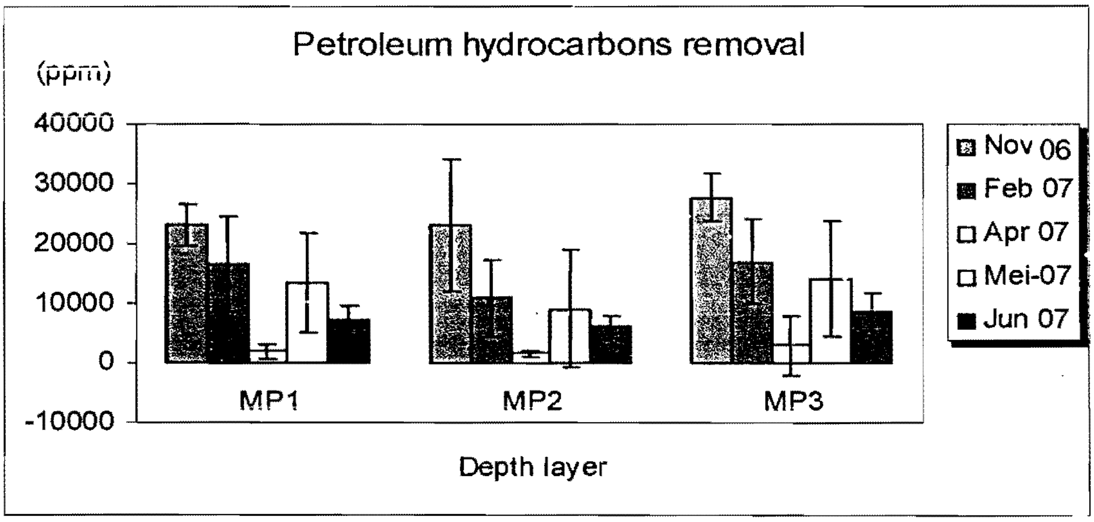

Figure 3. Petroleum hydrocarbons removal in mud pit

Note: MP 1: Sampling point at 0-30 cm of depth, MP 2 : (75-100 cm), MP 3 (> 150 cm). Indicated bars are average from 5 independent sampling points. (*i.e.* 1, 2, 3, 4, 5 which represents east-, south-, west-, north- and center of mud pit side respectively).

As far as bioremediation process in mud pit (Figure 3) shows the concentration of TPH decreased gradually during 5 months of treatment. As it can be seen, on May 2007, TPH levels are still high. The anomaly of such condition can be explained by the facts that the existing mode of operation to gain perfect mixing was not sufficient to create homogenous condition where some spots of contaminant concentrations were more elevated in certain areas compared to others. As a result, the bacteria used in the remediation techniques needed more time to lower down the pollutants.

Since the biodegradation is dependent on specific organisms' enzymes. The microbial tends to have optimum pH between 6.5 and 7.5, which equals to the intracellular pH. The pH will decrease due to microorganism's activities, however pH are measured and adjusted with lime addition periodically.

4. Monitoring Wells

Two monitoring wells were installed into the mud pit for leachate identification. The water was analyzed on pH, Electrical Conductivity and TPH. Preliminary survey shown the electrical conductivity in mud pit (water, slurry and solid) was 2002 ± 806 μmhos/cm, high cation contribute from heavy metal. After the whole sludge has been treated (eight months working period), the average of the electrical conductivity was 448 ± 175 μmhos/cm. The water wells analysis result shown average on Hg, Pb, Cd, Cr, Ni, Co, Fe, and Cu were less than 0.0002 ppm, 0.16 ppm, 0.001 ppm, 0.01 ppm, 0.05 ppm, 0.04 ppm, 0.11 ppm, and 0.11 ppm respectively. This result confirmed that no potential leach from bioremediation process. Suspect high level of EC may probably come from other ionic cations such as Na^+, K^+, Ca^{++} and, Mg^{++}.

5. Acute Toxicity Test (LD_{50}; Lethal Dose)

The results shown no significant clinical change and mortality of test animal until the concentration reaching 23,866 – 27,748.9 ppm. However the treated soil can not be exposed out of control when referring to the LD_{50} regulation (PP No. 74/2001) with 15.000 mg/kg body weight. A chosen to land spread the soil within perimeter was the best option.

End-Point Criteria and bioremediation monitoring

According to the Government of Indonesia standard KepMen LH No. 128/2003, end-point criteria of Bioremediation process is to achieve TPH, PAH (*PolyAromatic Hydrocarbon*), BTEX (Benzene, Toluene Ethyl Benzene and Xylene) concentrations and metals content shall meet threshold limits where the TPH should less than 10.000 ppm, BTEX and total PAH should not excess 10 ppm, except for benzene (1 ppm). From the the whole treatment process lagoon, slurry and biopile, the concentration of BTEX and PAHs were not detected.

Total Characteristic Leachate Procedure analysis for nine heavy metals such as Arsen, Barium, Cadmium, Chromium, Cooper, Lead, Mercury, Selenium and Zinc should not excess 5, 150 ppm, 1 ppm, 5 ppm, 10 ppm, 5 ppm, 0.2 ppm, 1 ppm, 50 ppm respectively. Once the treated soil has reached the end-point criteria, TPH concentration should be monitored every six months, for two years.

To validate the achievement of end-point criteria, we have conducted three times of monitoring on bioremediated soil from land spread area (each three months).The results showed on Table 2 After one year monitoring program, the average of TPH of bioremediated soil was below 2150 ppm.

Table 2. Monitoring of TPH Concentration

No	Sampling location	Depth (cm)	Monitoring Test Result of TPH (ppm)			
			Oct-07	Jan-08	Apr-08	Ags-08
1	Ex-Mudpit and Process	0-20		769	1260	1050
		40-80		ND	2410	1970
		0-20		ND	1250	ND
		40-80		392	461	1060
2	Land-spreading 1	0-20		3095	1800	704
		40-80	1160	3440	475	3690
		0-20		2610	2900	807
		40-80		2290	3550	1060
3	Land-spreading 2	0-20	1800	2460	3550	3970
		40-80	3550	642	1650	4760
		0-20		2890	872	2970
		40-80		1560	1050	594
4	Land-spreading 3	0-20		3850	3070	2940
		40-80	2750	3570	733	3460
		0-20		4090	1980	2490
		40-80		1170	3190	2500
	Average (AVG)		2315	2057	1700	2129
	Standard deviation (STDEV)		1051	1401	1044	1417

ND = Not detected (Detection Limit of 40 ppm)

The bioremediated soil was spread and planted with sun flower, legume cover crops (LCC), Jatropha, and wild plants that all plants grown healthy and fruity well. During first monitoring on January 2008, through visual observation, we suggested more than 80 % of land-spreading area was covered by the wild life habitats including grass, legume, shrub, and trees (Table 3). Some wild animals such as birds, snake and biawak were found (Table 3). The use of Velvet bean, Sword bean, Physic nut and Bellyache bush for polishing treatment post bioremediation have been reported. The potential use of some grass and LCC can be applied for revegetating because of they should be at least pre existing at site (local indigenous plant), tolerant to contaminant and may posed root architecture in density and depth. Through such potential species , they could provide the monitored reduction of remaining degradable hydrocarbons in soil, and leachate control, revegetating make aesthetic ecologically, therefore restoring the soil. On August 2008, the visual observation indicated that almost 80% land spreading area 1, 2 and 3, and ex mudpit. were covered by many plants mention as reported on January and April 2008. The tree of Jatropha, papaya and cherry grew well up to 3 meter heigh.

Table 3. Biomonitoring on Land Spreading Area.

Wild life (local/common name)	Proximate identification
Grasses/poales	
Alang Alang	*Imperata sp*
Grass Pea	*Lathyrus sp*
Rumput Gajah	*Penisetum purpureum*
Rumput Kerbau (Bahia grass)	*Paspalum sp*
Rumput Kipas	*Selaginella sp*
Rumput Kuda (Bunch grass)	*Panicum sp*
Rumput para	*Brachiaria sp*
Rumput parit	*Axonopis*
Leguminosaea/shrub/herb	
Legume	*Sytlosanthes*
Putri malu (Share plant)	*Mimosa pudica*
Rice (Padi)	*Oryza sativa L.*
Sword bean	*Cannavalia gladiata*
Velvet bean	*Mucuna pruriens*
Etc..	
Plants	
Ubi jalar (Cassava)	*Manihot utillissima*
Jarak pagar (Physic nut)	*Jatropa curcas*
Jarak ulung (Bellyache bush)	*Jatropa gossypfoli*
Kersen (Singapore cherry)	*Muntingia calabura*
Papaya	*Carica papaya L*
Pisang (banana)	*Musa sp*
Sengon butho	*Enterolobium cyclocarpus*
Etc...	
Animals	
Ular tanah (Snake)	Calloselasma sp
Biawak air	Vananus salvator
Wild birds	Unidentified presence

IV. CONCLUSION

The Environmental Management Program, ExxonMobil Oil Indonesia Inc., (EMOI) Aceh Production Operation area conducted a bioremediation project to clean-up the existing mud pit at Cluster IV Arun field to meet the regulatory requirement limits. The characteristic of mud pit contained three phases of water, slurry and sludge. The characteristics of sludge showed that the Total Petroleum Hydrocarbon (TPH) concentration was in the range of 2-4% and heavy metal compounds such as lead, zinc and mercury were above the government regulation limits.

Three different biological treatment methods such as biopile, water lagoon and slurry were applied to reduce TPH pollutants from mud pit. The results shown biopile method was effectively removed more than 90 % of TPH in one month of treatment. Water lagoon method, was observed enable to reduce several of regulatory key parameters to meet the standards, such as oil and grease, pH, COD, H_2S, NH_3, and total Phenol. While the result from slurry method were effectively worked with direct application into the mud pit. Combination of those methods resulted in an effective and efficient treatment to reduce TPH as well as to meet the government regulatory standard for bioremediation. The environmental significance of this bioremediation project was successfully demonstrated by acute toxicity test (LD_{50}) to *Mus Musculus* that showed significantly improved sludge quality.

The bioremediated soil was spread and revegetated. The plants apllied were sun flower, legume cover crops (LCC), Jatropha, and wild plants that all plants grown healthy and fruity well, After one year monitoring program, almost 80% land spreading area were covered by those plants, the average of TPH of bioremediated soil was below 2150 ppm, the water quality of two monitoring wells were below regulation limit, even the electric conductivity tend to increase gradually.

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