

## A Field Scale Study of Serial Treatment using Slurping and *in situ* Soil Flushing to Remediate an Oil Contaminated Site

HWAN LEE<sup>†</sup> and YOON-JIN LEE\*

Department of Environmental Engineering, Cheongju University, 36, Naedok-dong,  
Sangdang-gu, Cheongju, Chungbuk 360-764, South Korea  
E-mail: yjlee@cju.ac.kr

A combined treatment of bio-slurping, to extract soil vapour and withdraw free-phase oil and *in situ* soil flushing, to remove residual pollutants distributed in the saturated zone, was performed for 9 months and 1 month, respectively, to remediate an oil contaminated site that covered an area of 1,500 m<sup>2</sup>. Twenty-eight extraction wells were used for the operations and average monthly amounts of soil vapour extraction were 385,000 m<sup>3</sup>. The removal efficiency for total petroleum hydrocarbons (TPH) and BTEX compounds (benzene, toluene, ethylbenzene, xylene) was 59.5 and 79.8 %, respectively, and was achieved after applying the two processes over a 10-month operational period. The spread of free-phase oil contamination followed the direction of ground water flow in this area. Removal of oil was less successful at higher ground depths. Concentrations of TPH and BTEX decreased by 64.1 and 87.1 %, respectively, at a subsurface depth between 6 and 7 m following the application of these two processes.

**Key Words:** Total petroleum hydrocarbons, Surfactant, Bio-slurping, *in situ* soil flushing, Remediation.

### INTRODUCTION

Soil, along with water and air, is a basic component of the earth and has a crucial role in material cycling. Grasping of the pollution extent, remediation and monitoring are difficult<sup>1</sup>. In addition, soil pollution can cause deterioration of ground water and can affect public drinking water supplies. Restoration of polluted soil is difficult with the short-term remediation process for the already polluted area in field and soil itself characteristics. A long-term solution at high expense is generally needed for the treatment of polluted soil and groundwater. This can involve the combination of several treatment processes because restoration may not be efficiently achieved with a single technology.

Energy consumption increases with the rise in the number of vehicles and industrial activities. However, petroleum is regarded as a main source of soil pollution, particularly during the processing and storage steps and this issue has been attracting

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<sup>†</sup>Department of Civil, Urban and Geosystem Engineering, Seoul National University, Gwanak 599, Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea.

more attention<sup>2,3</sup>. A large amount of oil is lost during exploration, production, refining and transportation, resulting in negative impacts to the environment<sup>4,5</sup>. In Korea, sources of petroleum pollution include gas stations, the trans-Korea pipeline (TKP) and military facilities, and these are also likely to be the sources of petroleum contamination to the soil<sup>6</sup>. Petroleum leaks and petroleum by-products, often referred to as non-aqueous phase liquids (NAPLs), can occur from underground storage tanks in industrial facilities and gas stations. Once in the groundwater and vadose zone<sup>7</sup>, non-aqueous phase liquids can contaminate water supplies for long periods of time and over an extensive area due to pollution migration with the movement of ground water.

Various technologies are currently being applied to remediate petroleum-polluted soil. Pumping methods include soil vapour extraction, bioventing, pump and treat and air sparging<sup>2,8-10</sup>. Bioslurping is a technology that combines soil vapour extraction and vacuum-enhanced pumping in order to recover free product from ground water and soil at petroleum-release sites that have light non-aqueous phase liquids (LNAPL) pollution<sup>11-13</sup>. Bio-slurping is advantageous at sites having medium to coarse-grained materials and in fractured rock<sup>10</sup>. This method is preferred to address oil contamination that occurs in boundary areas between groundwater-saturated and groundwater-unsaturated soil<sup>14</sup>. Compared with other remediation processes, bio-slurping is reported to be cost effective<sup>10</sup>.

Pump and treat, which has conventionally been applied for the remediation of soil and groundwater, is not as effective in removing NAPLs<sup>15</sup>. Relatively long treatment periods are required compared to other technologies and a high expense is incurred because NAPLs are relatively insoluble in water. Consequently, surfactant applications have been evaluated to address this. Soil flushing with surfactant is considered a faster, more economical method for soil remediation<sup>16-19</sup>. During a first flush of diesel polluted soil, 98.9 % of BTEX was removed using calgonite 1 %, which is 2.7 times more compared to using only water<sup>20</sup>. Choi *et al.*<sup>15</sup> reported an enhanced efficiency of mixed surfactant with polyoxyethylene (POE) and sodium dodecyl sulfate (SDS) on petroleum polluted soil, with the highest TPH removal occurring when these substances were at a 1:1 ratio. Washing rate with SDS recovered from effluent discharge was higher efficiency than that of initial SDS<sup>21</sup>. Using surfactants to enhance the solubility of NAPLs has been investigated as a technique for soil remediation. The efficiency of using surfactants depends on the characteristics of the surfactants, the matrix and the pollutant.

*Ex situ* techniques are generally regarded as being more costly in terms of environmental effects and have limited applicability<sup>22</sup>. *In situ* soil flushing, which is a modified method of pump and treat, can remediate both soil and ground water in polluted areas without environmental alteration<sup>23</sup>. This process does not require excavation or the transportation of hazardous material because soil flushing is performed *in situ*<sup>10</sup>. Contaminated soils are flooded using injection wells and pollutants in the soil solution are transferred to a site where they can be removed<sup>24</sup>, captured

and pumped to the surface using extraction wells. This method can be applied to all types of soil contaminants, such as volatile organic compound (VOC), fuel, pesticides and inorganics.

The objectives of this study were (1) to assess the effect of soil remediation using the combined treatments of slurping and *in situ* flushing with surfactants in a fuel-polluted area in Korea and (2) to evaluate the variations in performance of these processes over a 10-month period. This was the first time that the combined treatment of slurping and *in situ* flushing on a field scale has been tried in Korea to remediate an oil polluted military site. The removal efficiency for the oil pollution area reduction under legal regulation and levels of TPH and BTEX was investigated during running this plant. The flushing effect was evaluated at different doses and types of surfactant were evaluated to determine an optimum surfactant for the field conditions. The reduction in thickness of the free-phase layer was enumerated after 9 months of using bioslurping. This research included that the availability evaluation on a field scale for the combined treatment of bio-slurping and *in situ* flushing and surfactant usage during soil flushing to remediate an highly oil contaminated site.

## EXPERIMENTAL

The target area was situated in a military base that had been polluted primarily with gasoline and diesel oil. The first signs of leakage appeared *ca.* 10 years ago, but it was not identified until excavation work in 2004 for the plumbing of a new oil pipe line that the extent of the leakages were determined. A primary source of petroleum contamination at the site was identified as being leaks from a deteriorating oil pipe line ramified from ground tank by preliminary investigation. The pollution covered an area of 1,500 m<sup>2</sup> to a subsurface depth between 6 and 9 m. Analysis showed the mean level of TPH in the target area to be 3,010 mg/kg, which was 1.5 times over the restricted level<sup>25</sup> as specified by the standards for soil conservation in Korea. The average level of BTEX was 397 mg/kg, which was 5 times higher than the Korean standard for the conservation of soil<sup>25</sup>. The geological stratum for the target area was composed of sand to a subsurface depth of 6-7 m and a weathered layer of silty sand below 7 m.

Bio-slurping was initially applied to extract volatiles in the soil and recover petroleum and this was followed by soil washing, which successively removed residual pollutants in the saturated zone. This system is connected to 28 wells, shown in Fig. 1. The radius of influence for 23 wells for which bio-slurping was applied for free-phase oil extraction and vapour extraction was 5.5 m, while the radius of influence for 5 wells for which polluted water was pumped out was 7.5 m. The purification unit was composed of various equipments, including that for vapour extraction, chemical injection, air jetting system and ground water purification in a steel container (Fig. 2). The thickness of the petroleum layer and ground water level was measured weekly, using a Solinst 122 model interface meter. The installed system was operated 12 h each day, 5 days per week.

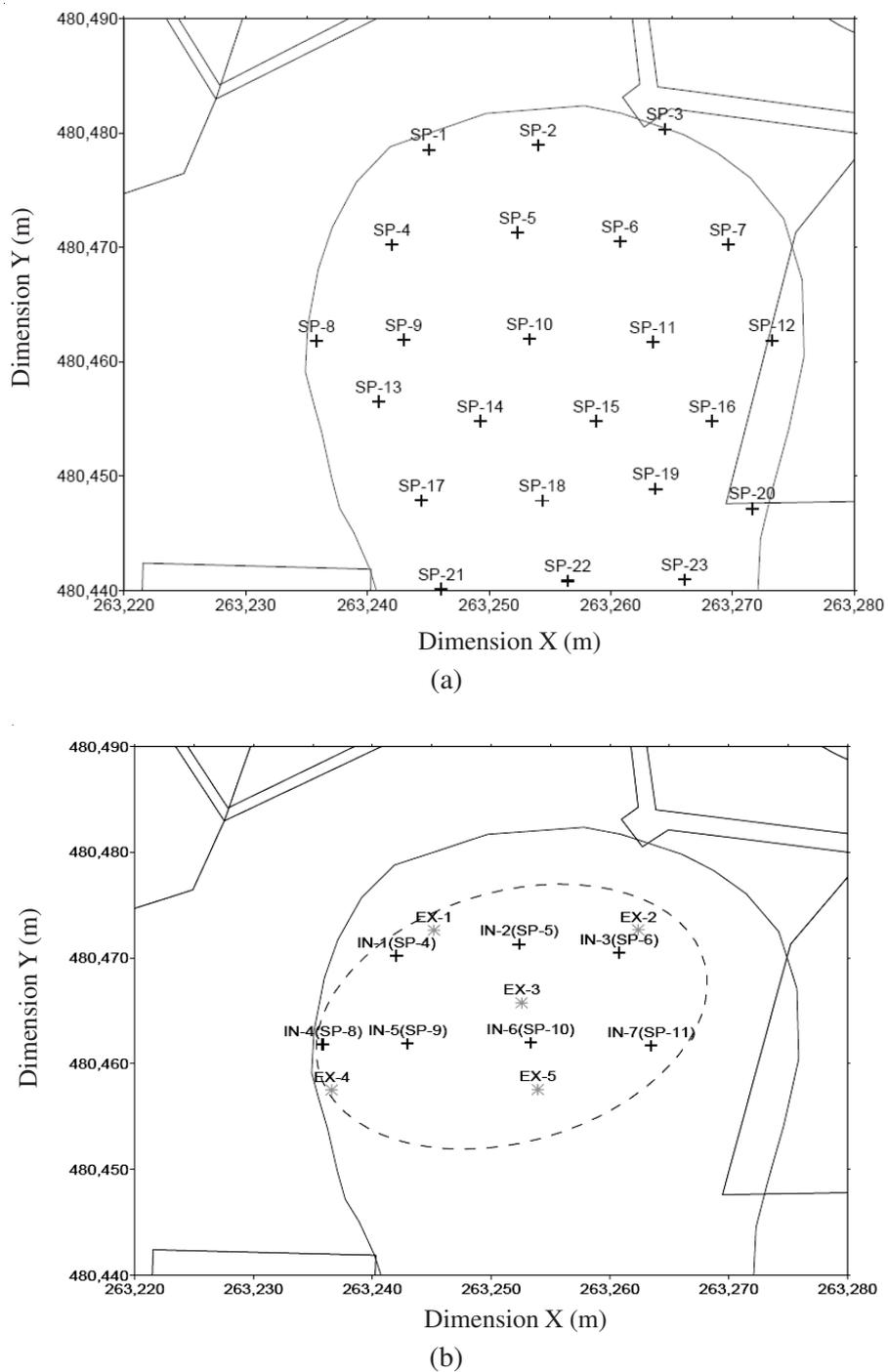


Fig. 1. Distribution of installed wells for the application of bio-slurping (a) and *in situ* soil flushing (b) within this the military base



Fig. 2. Photograph of the installed soil purification system

The wells were constructed with 100 mm diameter PVC pipes to a subsurface installation depth of 9 m, with the section between 5 and 9 m being constructed with perforated pipes. Water pumping and soil vapour extraction was 400 tons and 385,000 m<sup>3</sup> per month during the period of operation. The extraction of vapour per well was 1.06 m<sup>3</sup>/min and the extraction ratio of vapour:liquid ranged between 0.19 and 4.76 m<sup>3</sup> of air/L of water. The applied vacuum ranged between 0.25 and 0.38 bar, depending on the characteristics of the connected wells.

The soil particle distribution for this polluted area was: sand 70.7 %, silt 15.8 %, and clay 13.5 %, which corresponds to sandy loam by a soil texture calculator of the U.S. Department of Agriculture (USDA)<sup>26</sup>. Oil polluted soil was analyzed in the laboratory for TPH and BTEX. Soil smaller than 2 mm was separated through a sieve and agitated for 1 day at room temperature. The washing function of two surfactants, Tween 80 (sorbitan monooleate; Samchun Chemicals) and sodium dodecyl sulfate (Junsei Chemical), was compared in order to select the optimal surfactant for soil flushing. Surfactant, at concentrations of 0.5, 1.0 and 2.0 %, was added to a soil sample in 500 mL beakers. The solutions were agitated in a jar-tester at 130 rpm for 2 h.

Total petroleum hydrocarbon level was analyzed using the standard method for soil in Korea<sup>27</sup>. Briefly, 10 g of soil is mixed with 30 g of anhydrous sodium sulfate in a bottle and then dichloromethane is added. Contaminants were separated by ultrasonic extraction and the extraction filtrate was percolated through a 4 µm membrane (adventec 5B) and then measured by gas chromatography (HP 6890) with a flame ionization detector (FID).

Sampled soil (5-10 g) for the determination of BTEX was preserved with 10 mL of methanol in 30 mL glass bottles and transferred to the laboratory where they were kept at a temperature between 0 and 4 °C. The same amount of anhydrous sodium sulfate was added after placing 150 µg of the internal standard material in the bottles. This was mixed by shaking for 2 min and stationed. The upper 2 mL was collected after centrifugation for 3 min at a relative centrifugal force over 150, and it was determined by GC-FID with purge and trap.

## RESULTS AND DISCUSSION

**Application of bio-slurping:** The distribution of contaminants can be affected by rainfall fluctuations because this area is an unconfined aquifer. We therefore measured rainfall to determine seasonal variation (Fig. 3). Yang and Ahn<sup>28</sup> reported that ground water level varies according to the adjacent surface water level and rainfall penetration process.

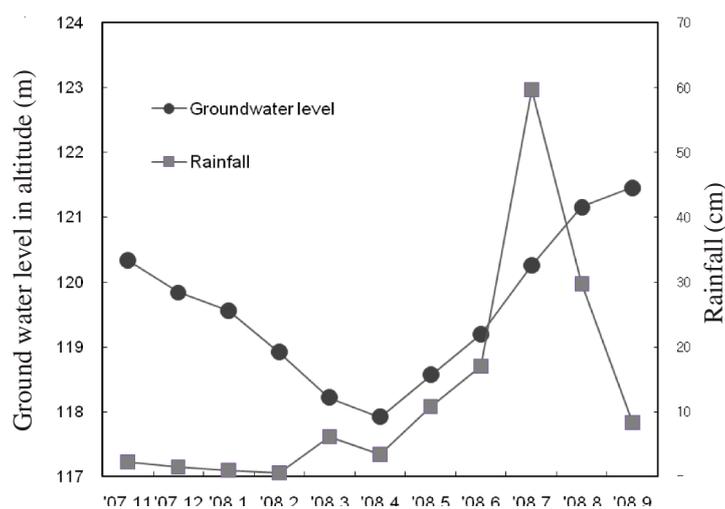


Fig. 3. Variation in monthly average rainfall and ground water level

Water level in altitude varied between 117.9 and 121.2 m during this experiment. The light non-aqueous phase liquids layer possibly moves with the variation in water level and could contribute to pollution migration over up and downside into soil. Ground water levels gradually decreased from November 2007 to April 2008, with the lowest value recorded at 117.9 m in April 2008, before increasing until September 2008. These results are consistent with results from the Gum river in Korea<sup>28</sup> where relatively high levels of ground water were maintained through the summer and early fall and lower levels occurred during the winter and spring.

The existence of free-phase pollutants and the distribution of contaminants in the saturated/unsaturated zone with a varying ground water level are important considerations when using the soil flushing technique. Bio-slurping was used initially

while a high level of free phase layer exists and unsaturated zone pollutants increase according to low water level. The recovery of free-phase oil and vapour extraction by bio-slurping occurred over a 9 month period. This was followed by *in situ* flushing has operated after August, which the thickness of free phase oil evidently reduced and pollutants in saturated zone increase with high water level.

Bio-slurping resulted in a reduction in the thickness of the petroleum layer over time (Fig. 4). The maximum and mean thickness levels were 45.2 and 72.0 % in December 2007 compared to those of November 2007 after the bio-slurping operation. The maximum and mean oil thickness levels at November 2007 (163 and 10.7 cm, respectively) decreased to 7.0 and 0.1 cm, respectively, after 9 months of bio-slurping operations, corresponding to a 95.7 and 99.9 % efficiency, respectively. The application of bio-slurping technology efficiently achieved a significant reduction in the thickness of the free-phase layer in the study area.

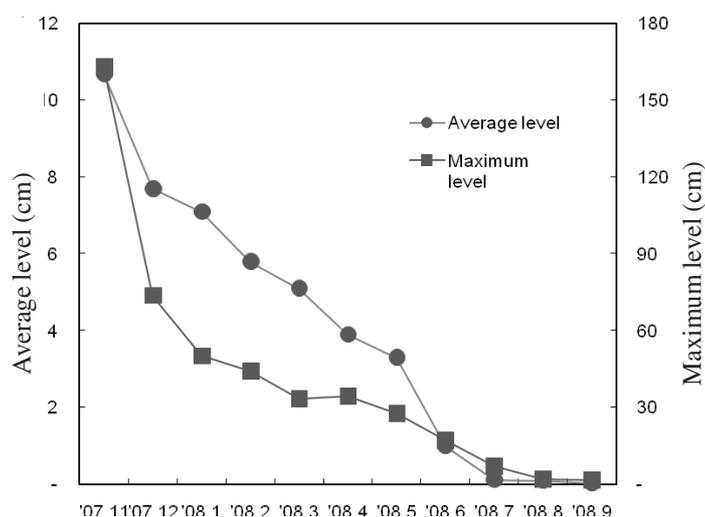


Fig. 4. Variation of free phase layer thickness on operation time

Gidarakos and Aivalioti<sup>13</sup> noted that groundwater table variations and certain meteorological conditions influence the efficiency of the bio-slurping process and the thickness of free-phase layer. It speculated that fluent oil substance had been preferentially removed in the upper portion of the ground water during bio-slurping and this could shorten the total purification period by preventing the additional spreading of oil pollutants during remediation. The slurping rate was reported to be slow at low temperatures<sup>10</sup>, but removal did not decrease during winter in present study.

Even with increasing ground water levels, free-phase petroleum, which is trapped within the pores of the silt layer at a depth between 7 and 9 m, could remain as residual LNAPL. Because the thickness of the free-phase oil could increase again during the dry season, continuous monitoring of the free-phase oil is necessary.

The relationship between ground water flow and the distribution of free-phase oil in the wells from December 2007 to May 2008 is shown in Fig. 5. Light non-aqueous phase liquids were distributed by ground water flow in a northwest to southeast direction. Free-phase oil having a thickness greater than 1 cm was primarily distributed in this zone. A high thickness between 10 and 35 cm was found in the center portion of this area, and free-phase oil was mostly found in the southeast part of the site. Kim and Yi<sup>29</sup> reported that horizontal spreading of LNAPLs was faster than that of dense non-aqueous phase liquids (DNAPL) after it reached on the water table and the LNAPLs form the continuous film on the water table and migrate horizontally with the flow of water in saturated zone.

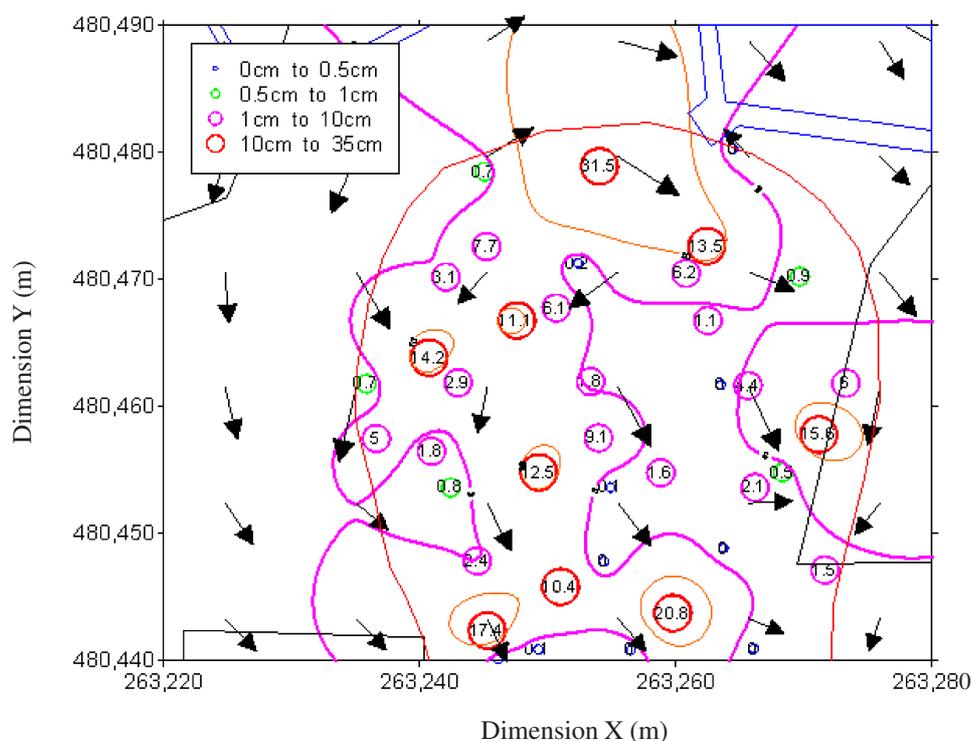


Fig. 5. Distribution of petroleum contamination and ground water flow in the study area

**Soil flushing:** The efficiency in removing TPH with the two surfactants is shown in Fig. 6. Sodium dodecyl sulphate (SDS) and Tween 80 were found to have a similar removal efficiency at 0.5, 1.0 and 2.0 % concentrations. This removal efficiency at a concentration of 0.5 % SDS and Tween 80 was 2.7 and 2.6 times higher, respectively, than that of water flushing (no surfactant). These results indicate that the use of surfactants is effective with *in situ* soil flushing. There was no notable difference in removal efficiency of TPH at surfactant concentrations between 0.5 and 2.0 %. Because the removal efficiency was highest for SDS, we decided to use

SDS at a concentration of 0.5 %, which was considered to be the most economical concentration, as the surfactant for *in situ* soil flushing. Lee *et al.*<sup>18</sup> reported that SDS was effective for the merit of the low possibility for the second pollution occurrence in the surfactant flushing. The removal efficiency of the combined surfactants at a 1:1 ratio did not increase compared to a single surfactant in this experiment, which is different from the work of Choi *et al.*<sup>15</sup>.

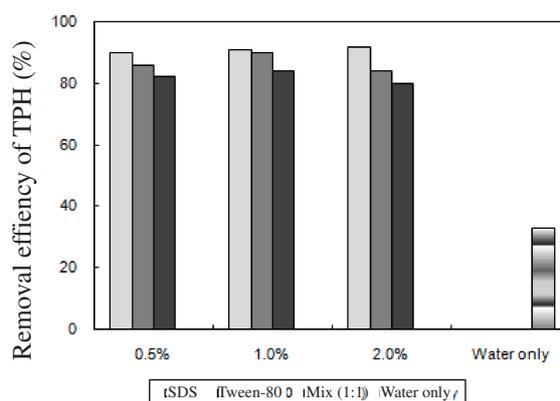


Fig. 6. Comparison of removal efficiency between the surfactants and water alone

The removal efficiency using 0.5 % SDS was evaluated in relation to soil diameter (Fig. 7). Total petroleum hydrocarbon concentrations were analyzed for soil particle sizes < 0.045 mm, between 0.045 and 1 mm and between 1 and 2 mm and were found to be lower with larger soil diameters prior to flushing. Following flushing, 100 % of TPH was removed from soil with particle diameters between 1 and 2 mm and the removal efficiency decreased as particle diameter decreased. The initial amount of TPH (14,000 mg/kg) was highest in soil particle sizes < 0.045 mm. Total petroleum hydrocarbon 8,000 mg/kg was removed after the application of 0.5 % SDS in soil particle sizes < 0.045 mm. FRTR<sup>30</sup> reported that treatment of heterogeneous soil by soil flushing is difficult because the flushing solution cannot move through a soil that contains a high percentage of silt or clay<sup>31</sup>.

*In situ* soil flushing started after August 2008, at which time the free-phase layer thickness had decreased considerably. In advance, soil flushing operation was performed for the highly polluted area 500 m<sup>2</sup> in the study area. Soil flushing took place for 22 days and simultaneously, pumping after the injection of SDA occurred through 5 neighboring points of the injection well. The daily application of SDA was 36.4 ton. Sufficient oil removal was difficult with the bio-slurping technology because residual pollutants were strongly adsorbed by the soil in the weathered layer. Soil flushing aimed to increase the solubility of the pollutants by injecting surfactant and separating the pollutants from the soil. The amount of ground water from the pumping well was 1,120 ton, which is 1.4 times than that of surfactant dose (Fig. 8).

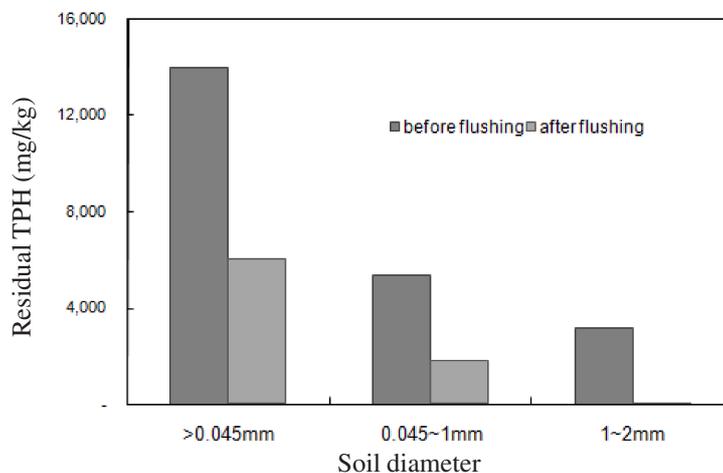


Fig. 7. Removal efficiency of TPH during soil washing in relation to soil particle size

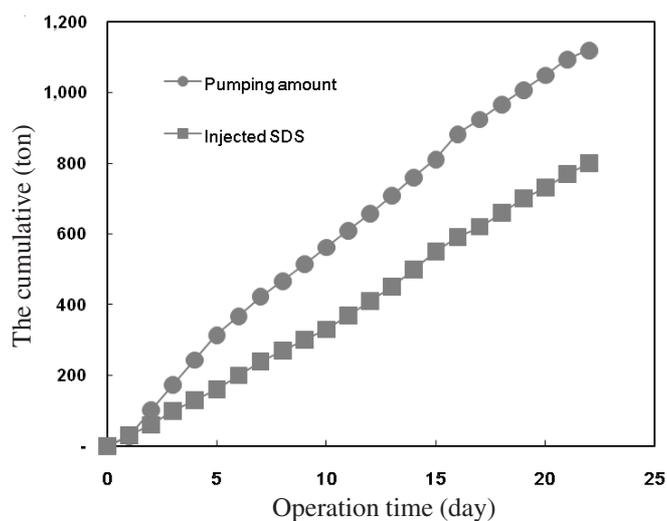


Fig. 8. Cumulative amount of SDS injected and pumping on operation time during soil washing

The variation in TPH levels at a EX 3 well is shown in Fig. 9. The initial TPH level in the ground water was 2.3 mg/L and these levels gradually increased over 8 days. Adsorbed pollutants on the soil were separated by *in situ* flushing. Total petroleum hydrocarbon levels then decreased with time after reaching a maximum concentration of 11 mg/L at 8 days. Considerable contact time should be needed to be attained the maximum value for the considering pollutants movement by specific channeling. It should be required significant times for the change of pollutant route with pumping and detachment of pollutants which exist at the high depth layer of saturation zone.

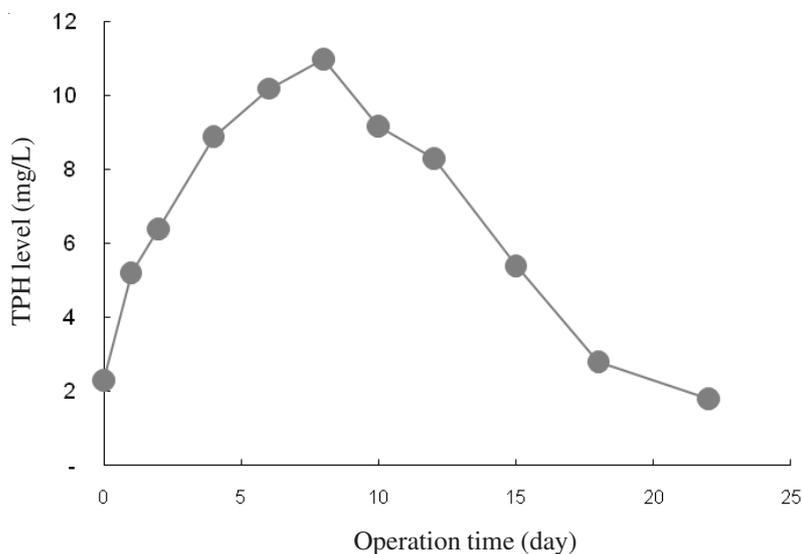


Fig. 9. Variation of TPH level at the well EX-3

The average reduction in TPH concentrations after *in situ* soil flushing in the 7 highly polluted points at a subsurface depth of 6-9, 7-8 and 8-9 m is shown in Fig. 10. The mean removal efficiency of TPH was 29.1 %. The removal rates at the different depths were 45.8, 17.9 and 15 % for 6-9, 7-8 and 8-9 m, respectively. The removal efficiency was lowest at a subsurface depth of 8-9 m, which is the weathered layer. The water permeability for the solid weathered layer was relatively low compared to that of the upper buried layer.

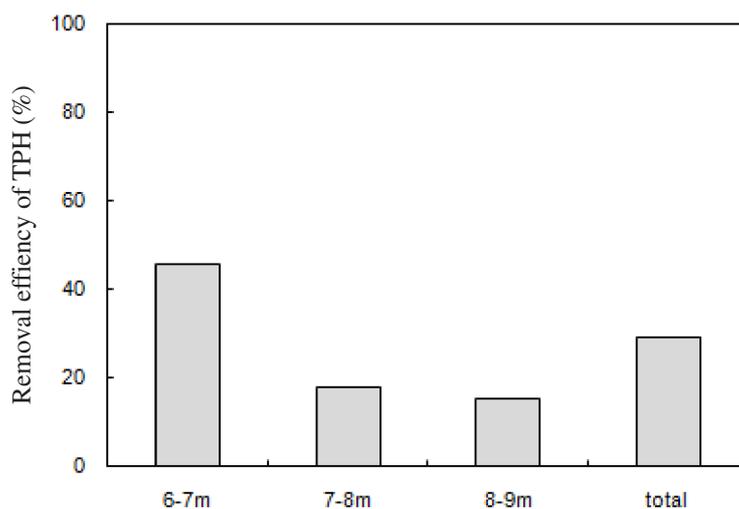


Fig. 10. Removal efficiencies of TPH after *in situ* soil flushing over a 500 m<sup>2</sup> area

The variation of polluted area over TPH 2,000 mg/kg on the subsurface depth was simulated in Fig. 11 during 10 month serial operation. In total, 59.5 % of the mean concentration of TPH was removed and 77 % of the TPH polluted area was remediated to a TPH concentration under 2,000 mg/kg, which is the legal standard, during the 10 month operation using bio-slurping and *in situ* soil flushing. The reduction efficiency of area of TPH with a concentration greater than 2,000 mg/kg was 86.8, 68.5 and 67.4 % for depths of 6-7, 7-8 and 8-9 m, respectively. The mean reduction in TPH concentrations following the application of both technologies was 64.1, 56.9 and 57.4 % for the subsurface depths of 6-7, 7-8 and 8-9 m, respectively (Table-1). The highest TPH removal efficiency occurred in the area between 6 and 7 m.

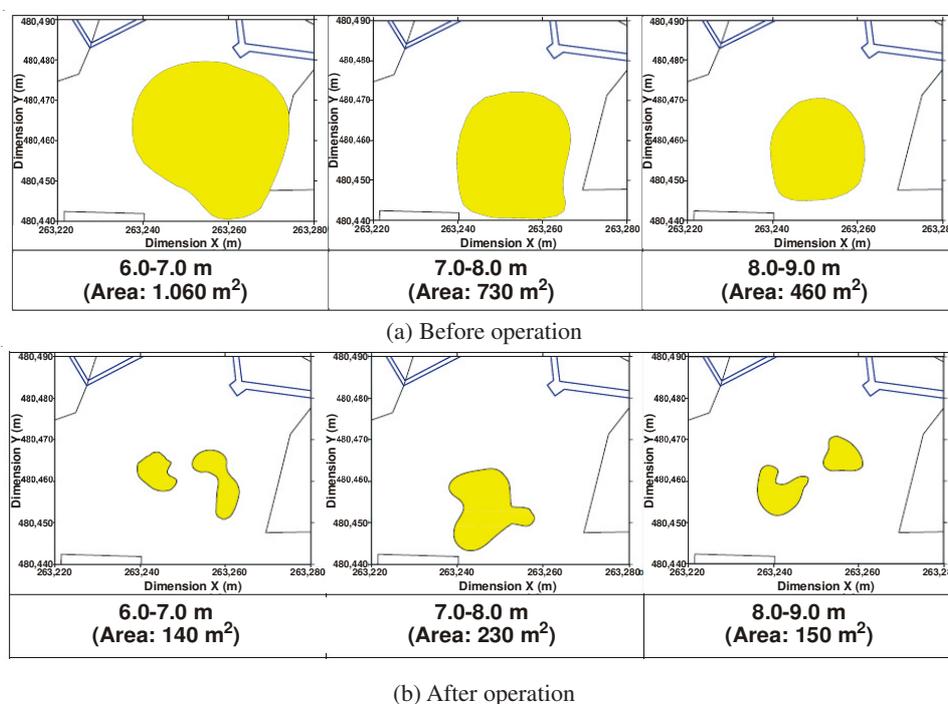


Fig. 11 . Comparison of pollution area over TPH 2,000 mg/kg between initial condition and 10 months operation later after the application of bio-slurping and *in-situ* soil flushing

TABLE-1  
TPH AND BTEX REMOVAL EFFICIENCY AFTER BIO-SLURPING AND  
SOIL FLUSHING IN RELATION TO STRATUM DEPTH

	TPH (%)	BTEX (%)
Subsurface 6-7m	64.1	87.1
Subsurface 7-8m	56.9	77.2
Subsurface 8-9m	57.4	73.4
Total	59.5	79.8

The variation in concentrations of BTEX in relation to subsurface depth during 10 months of bioslurping and *in situ* flushing is shown in Fig. 12. The polluted area (over 80 mg/kg) decreased from 3,610 m<sup>3</sup> to 1,090 m<sup>3</sup>. The area polluted by BTEX having a concentration greater than 80 mg/kg was 89.1, 64.1 and 41.7 % for subsurface depths of 6-7, 7-8 and 8-9 m, respectively. Removal efficiency for the mean level of BTEX was 87.1, 77.2 and 73.4 % for subsurface depths of 6-7, 7-8 and 8-9 m, respectively.

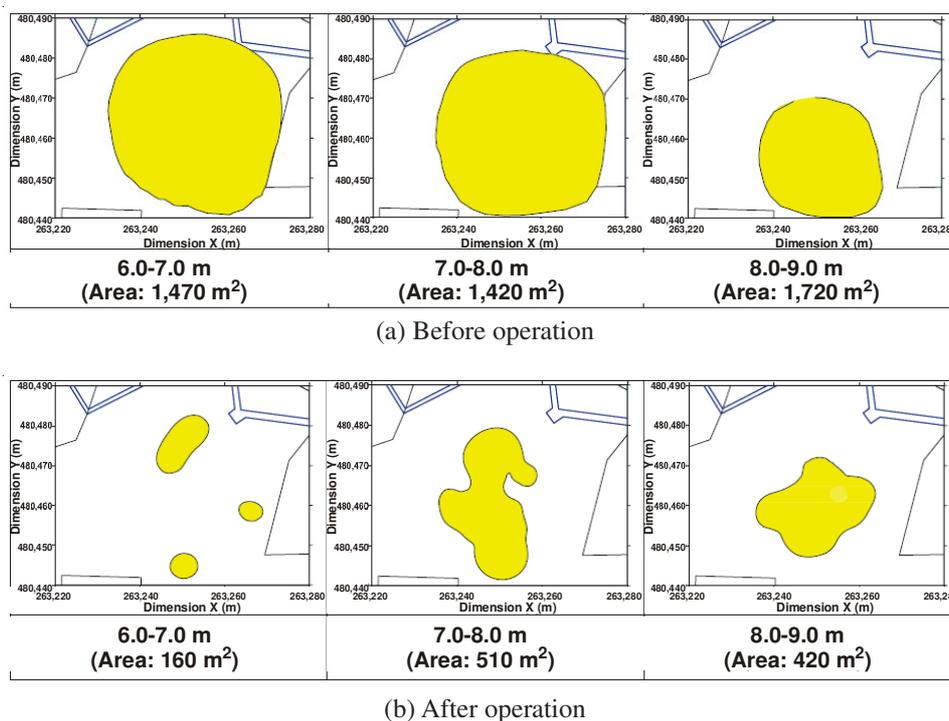


Fig. 12. Comparison between initial conditions and after 10 months of bio-slurping and *in situ* soil flushing of the polluted area where BTEX occurred in initial concentrations of over 80 mg/kg

Concentrations of BTEX were monitored throughout the operation period (Table-2). There was a reduction in BTEX concentration of 49.4 % during the first 3 months of bio-slurping and 79.8 % after 10 months. Initially, the maximum concentration of BTEX was 1,195 mg/kg. After 10 months of treatment, the maximum detected level of BTEX was 486 mg/kg, which was 6 times the mean concentration of 80 mg/kg.

The reduction in TPH and BTEX concentrations was efficiently achieved with bio-slurping and *in situ* soil flushing. However, the removal rate of TPH and BTEX in the weathered layer was shown to be relatively lower at subsurface depth compared to the upper layers. Enhancing the removal efficiency of a pollutant at a subsurface

TABLE-2  
VARIATION IN CONCENTRATIONS OF TPH AND  
BTEX OVER THE OPERATIONAL PERIOD

Operation period	TPH (mg/L)		BTEX (mg/L)	
	Mean level	Max. level	Mean level	Max. level
November, 2007	3,010	10,950	397	1,195
February, 2008	1,980	4,910	200	820
June, 2008	1,660	4,430	130	700
August, 2008	1,220	4,010	80	486

depth between 7 and 9 m would help in remediating a polluted area. A significant remediation effect was achieved with both processes. With a longer period of soil washing, an increased improvement in remediation would occur. Depending on the structure of the stratum, ground disturbance by physical methods and pollutant separation by high pressure air-sparging may be helpful in increasing pollution removal efficiency.

### Conclusion

This research evaluated the first continual application of bio-slurping and *in situ* soil flushing with a surfactant of an oil polluted military base in Korea. Soil vapour and oil extraction by bio-slurping was performed during the dry season when relatively high amounts of free-phase oil exist in the low ground water levels and this was followed by *in situ* soil flushing when high ground water levels were present.

Levels of ground water varied markedly over the year in the study area due to variations in precipitation. Bio-slurping decreased the thickness of the free-phase oil over the 9 month period from an initial mean level of 10.7 to 0.1 cm. The polluted area of free phase oil distributed which was similar direction with the flow of ground water.

The anion surfactant SDS was found to have the highest flushing effect of the two surfactants selected for this study and was applied at a concentration of 0.5 %, which was identified as being the most cost-effective. Removal efficiency for SDS decreased with a decreasing soil particle size. In contrast to previous published reports, the application of a combined surfactant did not show an enhanced flushing effect.

The removal efficiency of TPH and BTEX was 59.5 and 79.8 %, respectively, using slurping and *in situ* soil flushing with SDS over the 10 month operational period. The polluted area more than TPH 2,000 and BTEX 80 mg/kg, to levels within the guidelines for soil standards in Korea were reduced by 77.0 and 69.8 %, respectively. The reduction in oil was relatively less with higher subsurface depth levels. In conclusion, we recommend the extended operation of *in situ* soil flushing and the application of air sparging under high pressure for the removal of oil pollution in ground and the removal of residual oil adsorbed to soil particles.

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