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PART VII: Remediation

Chapter 14

***IN SITU* WASHING BY SEDIMENTATION METHOD FOR CONTAMINATED SANDY SOIL**

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ABSTRACT

We propose a new method of in situ soil remediation called in situ washing by sedimentation (IWS), accomplished by injecting a high air-pressure into a mixture of saturated water-sandy soil at a certain depth (D) and hydraulically separating the soil particles based on their particle size and density. This physical segregation exploits the distribution of contaminant in the soil by physically separating a selected contaminant-rich fraction. For the in situ application, the physical segregation by sedimentation and on-site water wash treatment happen as an integrated process. The advantage of IWS that the washing and segregation processes take place simultaneously during the remediation process, quick, effective and cheap since there are no costs for excavation of contaminated soil from the site. The effect of soil-water ratio and diameter geometry of the column on the effectiveness of segregation by IWS was investigated. A series of laboratory test were conducted to optimize the soil water ratio for the best segregation process. Soil-water ratio 1:2 (v/v) was found to be optimum for particle segregation produced by IWS . The suitability of IWS for Polycyclic Aromatic Hydrocarbon (PAH) remediation, such as Napthalene, Phenantrene and Pyrene, were examined by batch sedimentation column experiment. The laboratory experiment was effective to produce a distinct size segregation of the contaminated soil into the coarse and fine fractions, as well as the wash water, indicating that a significant reduction in Napthalene, Phenantrene and Pyrene level (90%) may be achieved. The experimental results show that the removal

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efficiencies depend on the initial concentration of PAH in the soil sample, the duration of washing and the addition of biosurfactant in the washing solution.

Keywords: In-situ washing, sedimentation method, soil-water ratio, metal contamination

1. INTRODUCTION

Several technologies have been developed for the remediation of contaminated soil. Broadly speaking, they can be grouped in the following categories, *ex situ* technologies where the contaminated soil is removed and treated away from the site that has to be cleaned up and *in situ* technologies, where the contaminated soil is cleaned on site.

There are various options in each of these categories. *Ex situ* technologies include incineration, extraction, *ex situ* washing, and *in situ* technologies include soil vapor extraction, soil flushing, bioremediation and phytoremediation. The major advantage of *ex situ* technologies is that it takes relatively little time to remove the contaminated soil. The soil is excavated, treated or replaced and then filled in again. By comparison with *in situ* methods, however, method of *ex situ* soil remediation has many drawbacks. They are expensive, large quantities of soil have to be transported often in residential areas, there are risks to buildings and other structures especially with major excavations, contaminants can be emitted during excavation, and it causes major disruption of daily life in the area to be remediated.

Increasingly, *in situ* technologies are used for sustainable clean-up of contaminated sites (Otten, et al., 1997). Air and water extraction have in practice proved to be reliable methods for various types of soil remediation (Bass et al., 2000, Reddy et al., 1995, Hutzler et al., 1991). Although bioremediation and phytoremediation was also used, this technology works too slowly to be fully effective as a remediation technology (Cunningham et al. 1995, Milic, et al., 2007).

Soil washing was conventionally performed *ex situ* in treatment plants that employ extracting chemical to remove contaminant from soil into aqueous solution (Mann, 1999; Abumaizar and Smith 1996; Cline and Reed, 1995; Griffiths, 1995). Few studies of *in situ* soil washing have been conducted, even though *in situ* soil washing could be suitable for certain contaminated soil in the field (Nash and Traver, 1993; Niven and Khalili 1998; and Makino and Kamiya, 2006).

We propose a new method of *in situ* soil remediation called *in situ* washing by sedimentation (IWS), accomplished by injecting a high air-pressure into a mixture of water-sandy soil column at a certain depth (D) and hydraulically separating the soil

particles based on their particle size and density, as shown in Figure 1 (Budianta et al, 2005, 2006a, 2006b). This physical segregation exploits the distribution of contaminant in the soil by physically separating a selected contaminant-rich fraction. Several researches indicate that the finest parts of soil are particularly active in the sorption processes of organic as well as inorganic contaminant (Evans et al., 1990; Hwang and Cutright, 2002; Abollino et al., 2003; Echeverria et al., 2002). For the *in situ* application, the physical segregation by IWS and on-site wash water treatment happen as an integrated process and it is important to isolate the site to protect the leakage of the aqueous solution used (Figure 1). The advantage of IWS was that the washing and segregation processes take place simultaneously during the remediation process, quick, effective and cheap since there are no costs for excavation of contaminated soil from the site. The fine fraction is recovered for further treatment or disposal. The wash water is completely collected, treated and recycled.

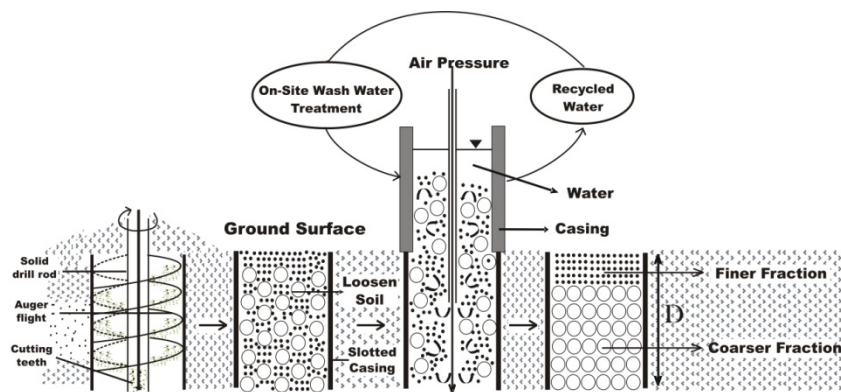


Figure 1. In situ Washing by Sedimentation Method (IWS)

The objective described in this paper, first was work on the simple-batch laboratory-column experiment of IWS by a systematic investigation on the evaluation of the effect of soil-water ratio and diameter geometry of column on the effectiveness of particle segregation by IWS in laboratory scale. Secondly, our second goal was to observe the removal of metal-contaminated sandy soil by IWS in batch laboratory experiment.

2. MATERIAL AND METHODS

Experiment were conducted on the uncontaminated soil were collected from Ota

District Tokyo in 1.5 m depths. The result of grain-size distribution indicated that the original soil sample contained approximately 10-20% clay-silt size particles and the remaining was sand (sandy soil).

2.1. The Effect of Soil-water Ratio on the Effectiveness of Particle Segregation by IWS

The objective of this experiment was to evaluate the effect of a soil-water ratio on the effectiveness of particle segregation by IWS. Environmental and economic concern required that the volume of water solution used on IWS in order to obtain sufficient particle segregation should be kept to a minimum. Generally, one of the main drawbacks of the washing method on soil remediation is the vast consumption of water required to make up the washing solution for the removal of the contaminants that have been retained in the contaminated soil. In IWS, we propose for washing solution which must be subsequently be on-site treated before it can be re-use. A series of laboratory experiment were carrying out to optimize the soil-water ratio for sufficient particle segregation.

Experiment was conducted in the cylinder tube of 2000 ml in volume with 90 mm inside diameter. For constant mass of 810 g dry soil, different volumes of water solution were used and were described in Table 1. The value of 300 ml volume of dry soil sample was divided by 2.710 g/cm^3 measured density.

After the soil sample and the water solution was prepared into the cylinder tube, the column then was stirred for one minute and let sedimentation occur for 30 minutes. Theoretically, the soil particle in the column will settle in a descending order of particle sizes with the top part of the soil layer consisting of smaller particle. The segregation of the soil particle into nominal size fraction in this experiment depends on the sedimentation process on hindered settling in the high sediment concentration.

Table 1. The water solution and the dry sample soil used

No. Tube	Soil sample gram (milliliter)	Volume of Water (ml)	Soil-Water Ratio (v/v)
1	810 (300)	300	1:1.00
2	810 (300)	400	1:1.33
3	810 (300)	500	1:1.67
4	810 (300)	600	1:2.00
5	810 (300)	700	1:2.33
6	810 (300)	800	1:2.67

The next step, all the cylinder tubes containing sedimentation soil column was kept in a refrigerator for -18°C temperature, in order to obtain an undisturbed frozen

soil column sample. After 24 hour, the frozen soil column samples then were taken out and marked. The purpose and the advantage of this freezing was to obtain a selected frozen column soil sample accurately by slicing the soil column without disturbing the sedimentation column. These undisturbed frozen soil column samples then were cut into several certain thicknesses and were analyzed for particle size distribution (Figure 2).

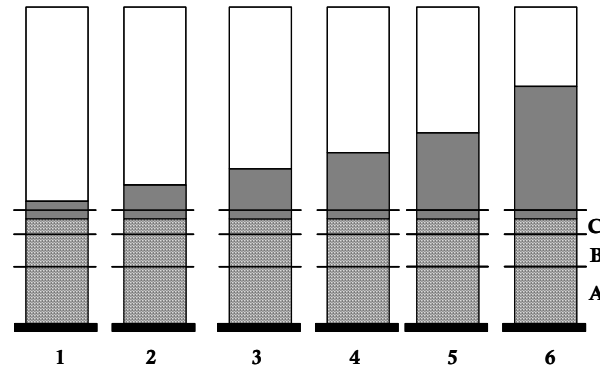


Figure 2. An illustration of undisturbed frozen soil column (*not to scale or actual size*)

The experiment was continued by observing more accurately the two soil fractions considered in this study. The terminology “fine” and “coarse” particles was used as the results of particle segregation by IWS. As previously stated, the top layer was assumed to be fine fraction and the bottom layer was assumed to be coarse fraction assumed reflects the high and low content contaminant in each fraction. Similar to previous experiment which conducted in the cylinder tube with 90 mm inside diameter, the optimized the soil-water ratio 1:2 (v/v) consist of 810 g dry soil and 600 ml volumes of water solution were used.

After sedimentation column was created, the top layer consisting of fine fraction was sampled very carefully by using small spoon, after the wash water was removed by suctioning. The fine fraction was then determined for particle size distribution analysis. Corresponding to the fine fraction, the remaining coarse fraction in the bottom part of the sedimentation column was also determined for particle size distribution analysis. The result of particle size distribution analysis was shown in Table 2. The fine fraction separated in the study was labeled as Clay and the coarse fraction was labeled as Fine Sand. These two fractions were then dried and weighed. The result can be seen in Table 2. The percentage of saturated volume was obtained by measured the height of each fraction in the sedimentation column.

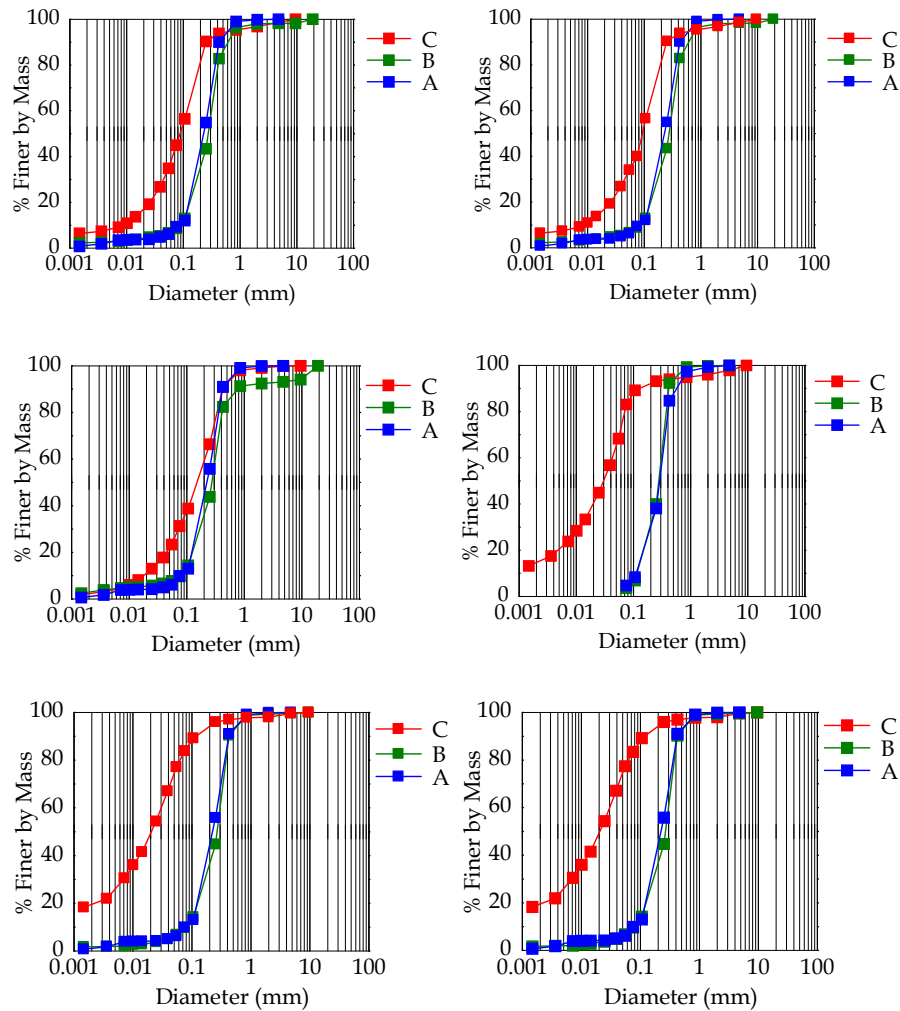


Figure 3. Particle size distribution analysis of selected frozen sedimentation column in each tube

In this stage, these two fractions separated (coarse and fine) was also determined for several parameter. It shows that the fine fractions have the high specific surface area, the high organic content, and containing of 1:2 clay minerals such as illite. It can be understood that the fine fraction separated by our sedimentation column particularly active in the sorption processes of contaminant.

Table 2. Particle size distribution analysis of fine and coarse fraction

Fraction observed (Size Diameter)	Fine	Coarse
Gravel 2-75 mm %	0.0	0.9
Sand 0.075-2 mm %	1.0	43.8
Fine Sand 0.075 mm %	6.9	48.5
Silt 0.005-0.075 mm %	37.0	5.1
Clay <0.005 mm %	55.1	1.6
Uniformity Diameter (mm)	-	2.59
	Clay	Fine Sand
Percentage Dry Weight	4.5	94.5
Percentage Saturated Volume	10	90

2.2. Removal of the organics contaminant on the sandy soil by IWS

2.2.1. Batch Sedimentation Column Experiment

Naphthalene (NAP), phenanthrene (PHE) and pyrene (PYR) were selected as example of PAHs representing organics contaminant. Two artificial contaminated soils were prepared by dissolving uncontaminated soil which collected from Ota District Tokyo (see Table 1 for detail) with an appropriate quantity of NAP, PHE and PYR solution as described by Sawada, et al., 2004. Briefly, uncontaminated soil sample was spiked with solution of NAP, PHE and PYR for three days to allow the dispersion and sorption of the contaminant in the soil matrix. (A) Soil with low concentration of PAH; (B) Soil with high concentration of PAH, by spiking uncontaminated soil sample with 500mg/kg of NAP, PHE and PYR solution for soil A and 1000 mg/kg for soil B. All samples then were determined for PAH concentration after ethanol digestion by using a Gas Chromatography Mass Spectrometry (GC-MS) under optimized operating condition. The resulting of the artificial contaminated soils had a final concentration of 30, 75 and 50 mg/kg of NAP, PHE and PYR on Soil A and 250, 490 and 350 mg/kg of NAP, PHE and PYR on Soils B.

Experiment was conducted by using air pressure created by air pump injected into the soil-water column on 90 mm internal diameter cylinder tube (Figure 4). A 0.5 kg dry PAH-contaminated soil sample was used and 370ml water was added based on the optimized soil-water ratio 1:2 (v/v) obtained from previous experiment. Seven similar tubes were constructed and the air pressure was introduced into each tube for 1, 2, 3, 4, 5, 10 and 15 minutes to observe the effect of washing duration.

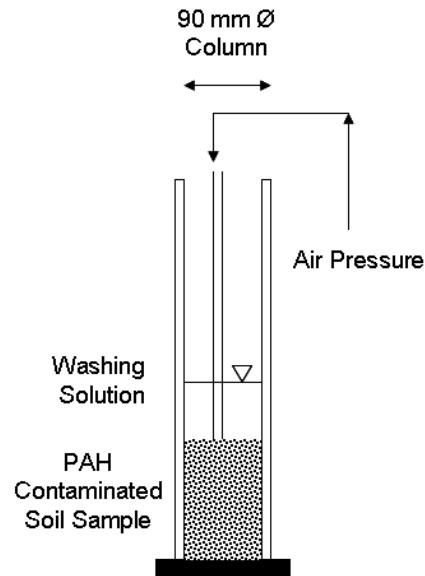


Figure 4. Laboratory experimental setup

The air flow started with the position of the pipe reaching the surface of the soil sample inside the tube. The air pressure rate was increased as the inlet pipe penetrated the soil sample. After the final depth was reached, air flow rate was kept constantly until the inlet pipe almost reached the bottom of the cylindrical tubes.

The soil inside the tube was allowed to settle to obtain clear water above the settled solids. The coarse fractions separated in this batch sedimentation experiment were analyzed for their particle size distribution. The wash water and the fine fraction were sampled through pipe by suctioning and the coarse fraction was sampled using a small spoon.

The segregation of coarse and fine fractions produced by IWS was investigated as a function of the washing duration, obtained by introducing air pressure into each tube for 1, 2, 3, 4, 5, 10 and 15 minutes. The result shown in Table 3 indicated that the result of segregation in our batch sedimentation experiment was reliable, and the accuracy of segregation increase depending on the washing duration. After 5 minutes washing, 92.2% of particles in the coarse fraction were separated as $>0.075\text{mm}$ particles size diameter (fine to coarse sand) and fine fraction separated as $<0.075\text{mm}$ particles size diameter (clay-silt). After 5 to 15 minutes, it seems that no significant difference occurred in the results. Perfect segregation was expected in this method, but it showed that only about 90% of its grains were separated.

Table 3. Values obtained by particles size analysis of coarse fraction separated by IWS

Sample (observed fraction)	Washing Duration	Sand % (0.850 mm-0.250 mm)	Fine sand % (0.250 mm-0.075 mm)	Silt % (0.075 mm-0.005 mm)	Clay % (<0.005 mm)
Original	0	42.1	33.7	12.8	11.4
Coarse	1	44.7	47.6	5.0	2.6
Coarse	2	44.8	47.5	5.0	2.5
Coarse	3	43.5	49.1	4.9	2.3
Coarse	4	44.8	48.2	4.7	2.1
Coarse	5	44.2	49.7	4.0	2.0
Coarse	10	44.8	48.2	4.7	2.1
Coarse	15	44.2	49.7	4.0	2.0

The inaccurate result is attributed by the lack homogeneity in the particle size distribution of the coarse fraction. The sedimentation process is more complicated if several particles are present and the system becomes a sediment suspension in hindered settling reflected when the concentration of the suspension decrease, the homogeneity of the separated fraction will increase and therefore impossible to exclude small amounts of finer particles. Although the result of segregation by IWS was not perfect, the first important point of this study was that the process succeeded to separate the soil sample into a coarse fraction and a fine fraction. The segregation into coarse and fine fraction will affect the high and low concentration of the contaminant in each fraction.

2.2.2 Removal of PAHs in Contaminated Soil

As shown in the result of the previous experiment, the accuracy of particle segregation was influenced by the washing duration, and consequently it will affect the percentage removal of contaminant. In this experiment, the coarse fraction sampled was a representation of a clean fraction.

Figure 5 shows the experimental data by using the deionized (DI) water and DI water with addition of biosurfactant as washing solution. In the case of DI water washing, the result show that the removal process rapidly reaches equilibrium, at approximately 10 minutes for each PAH; after this period no considerable changes in the removal rates were observed. The PAH in the contaminated soil sample was mostly must found in the wash water and the fine fraction.

The fine fraction in the seven tubes was collected, combined and analyzed for PAH concentration. The results of the GC-MS analysis on the fine fraction showed that the concentrations of the PAH were very high. The accumulations of PAH in the fine fractions are attributed to the high specific surface area, the presence of clay minerals such as illite, and the high organic content.

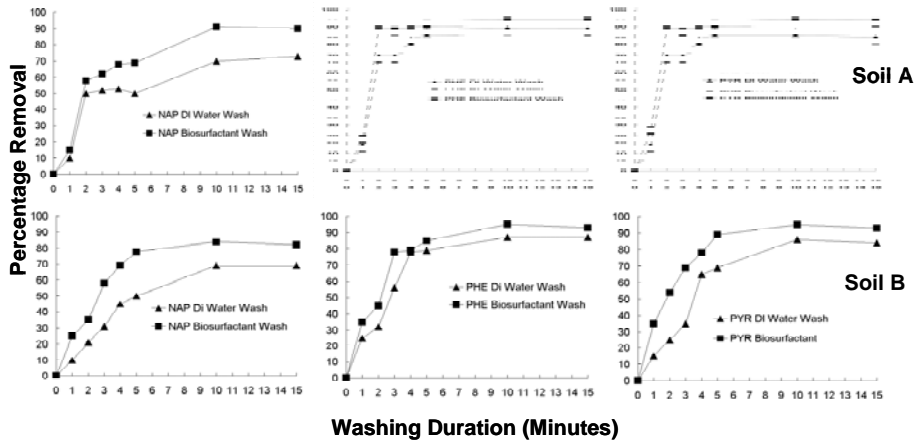


Figure 5. Percentage Removal PAH vs washing duration for soil A and B

The addition of biosurfactant (saponin) in order to enhance the percentage removal was also observed, by adding 0.25% by weight of saponin in the washing solution (Figure 5). In this stage, after fine fraction was removed, the wash water used was returned to the cylinder tube and saponin was added. The air pressure was then introduced into each tube for 1, 2, 3, 4, 5, 10 and 15 minutes to observe the effect of washing duration, the same as in the previous experiment. As shown in Figure 5, the removal percentage increased after the addition of saponin. The addition of saponin as an anionic biosurfactant was effective to assist in the solubilisation, dispersal and desorption of PAH from the contaminated soil fraction (Mulligan et. al., 2001).

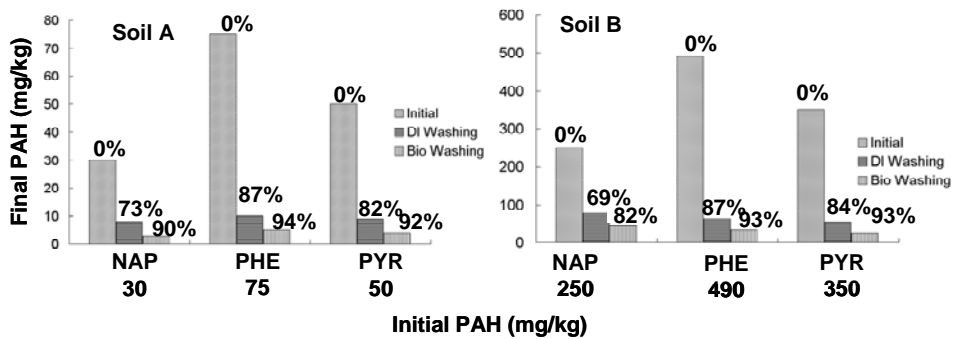


Figure 6. Removal efficiency of IWS on Soil A and Soil B

The result of the removal efficiency of PAH from soil A and B is shown in Figure 6. It shows that by using DI water only, approximately 80% of PAH retained in the

soil sample was removed. The addition of saponin has proven to be effective to enhance the removal efficiency up to more than 90%.

3. CONCLUSIONS

This study has addressed on the *in situ* soil remediation with an emphasis on the washing and segregation of soil particles by IWS. By using Ota District sandy soil as an object of this experiment, the following initial conclusions can be drawn from the results in this study:

The results of laboratory study on the effect of soil-water ratio on the effectiveness of particle segregation by IWS show that by using soil-water ratio 1:2 (v/v), the optimum for particle segregation on IWS was found. However, the result of laboratory study on the effect of diameter geometry of column on the effectiveness of particle segregation shows that generally the diameter geometry column will not affect the soil particle segregation.

The laboratory scale of in situ washing apparatus on IWS was able to produce a distinct size separation of the soil into coarse and fine and a significant reduction of Polycyclic Aromatic Hydrocarbon (PAH) such as Naphthalene, Phenanthrene and Pyrene level (90%) was achieved

The concentration of PAH contaminant was found to be a function of particle size; the coarse fraction were the cleanest and the fine fraction contained the highest PAH contaminant and a very small amount of the original contaminant was retained in the coarse fraction.

The removal efficiencies of remediation method proposed in this study depend on initial PAH concentration, the addition of biosurfactant in the washing solution and the duration of washing. Further treatment for the fine fractions and the wash water containing suspended solid particle and dissolve PAH contaminant need to be further investigated.

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