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Research Paper

Optimizing the on-site treatment of soil contaminated by heavy metals and petroleum using relocatable soil washing equipment

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ABSTRACT

In this study, the on-site treatment of soil contaminated by heavy metals and petroleum was tested using relocatable soil washing equipment for greater remediation efficiency. Different combinations of pH and solid/liquid ratio were tested to determine the optimum balance, settling on values of 5 and 1:2, respectively. Thereafter, soils containing Pb, Hg, and petroleum were further tested to assess the optimum number of washing cycles. The remediation efficiency of Pb and Hg in soil contaminated solely by heavy metals was 90.1 and 86.4% after three and two washings, respectively. The remediation efficiency of petroleum in soil contaminated solely by petroleum was 98.8% after one washing. When soil contaminated by both heavy metals and petroleum was cleaned, up to 91.0% of Pb, 86.9% of Hg, and 96.1% of petroleum were removed after two, one, and one washings, respectively. All remediation efficiencies and concentration reductions satisfied the standard threshold for soil contamination in South Korea.

Key words: Soil washing, remediation efficiency, relocatable soil washing equipment, on-site treatment, heavy metals, petroleum.

INTRODUCTION

Contaminated soil that needs remediation should be purified and/or transported, ideally being returned to a natural condition and setting (Adam et al., 2009). Ideally this would be done by immediate washing on-site, but this can be difficult to implement as environmental contamination is not always immediately detectable because soil contamination can proceed slowly (Huguenot et al., 2015). This can lead to widespread effects requiring purification efforts lasting decades.

In South Korea, the remediation of soil contaminated by petroleum is generally conducted by private companies using simple techniques (Ministry of Environment, 2010). However, soils in industrial complexes often contain complex contaminants, including heavy metals as well as petroleum, requiring simultaneous purification (Tang et al., 2012). Recent studies have focused on remediation through effective soil washing (Mohamed et al., 2013; Jelusic and Lestan, 2014; Zhang et al., 2015). Currently used equipment requires repeated treatments with excessive use of washing agent to reach remediation standards. Moreover, as this approach cannot purify contaminated soil on-site, transportation of soil to remediation facilities is required, which results in significant problems related to time constraints and environmental factors (Sabbas et al., 2003; Hyks et al., 2011) as well as economic costs (Kim, 2012). However, relocatable soil washing equipment can be moved rapidly to contaminated areas and directly purify contaminated soil on-site. In this study, the remediation efficiency of such equipment was verified and the optimum operating factors for remediation of soil contaminated by heavy metals and petroleum was evaluated.

MATERIALS AND METHODS

The tested soils were contaminated solely by heavy metals (HM), solely by petroleum (P), and by a combination of both (HM+P). The sampling site was a typical industrial complex

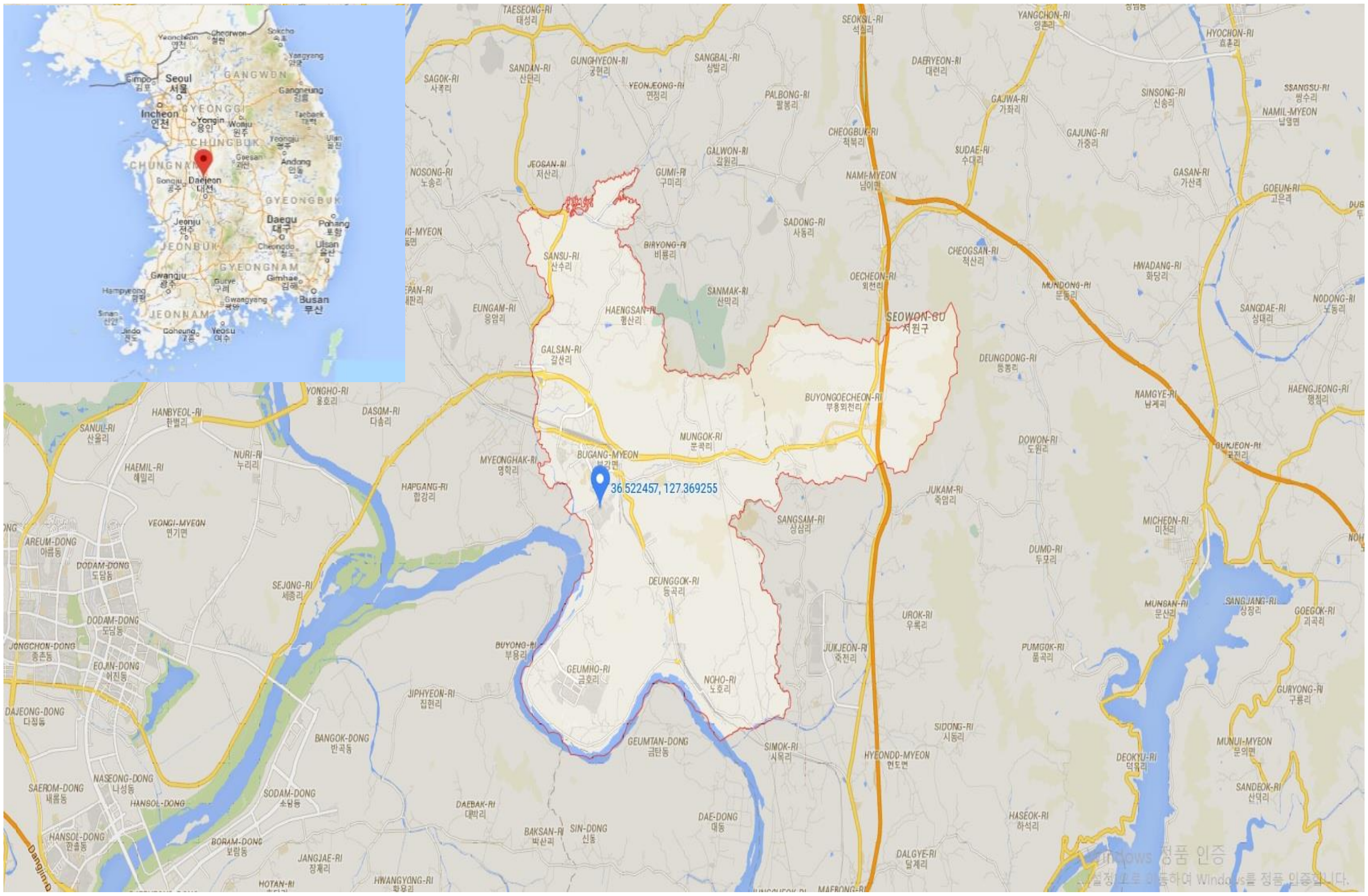

Figure 1: Location of sampling site in central South Korea (see Figure S1 for further detail).

Table 1: Average contaminant levels in tested soils.

Item	HM*	P*	HM+P*
Pb (mg kg^{-1})	1,222.3	-	1,156.1
Hg (mg kg^{-1})	570.7	-	221.6
TPH (mg kg^{-1})	-	5,222.9	6,127.2
Contaminated area (m^2)	5,275	1,532	3,872
Contaminated quantity (m^3)	6,582	2,234	6,217

*HM (soil contaminated solely by heavy metals); P (soil contaminated solely by petroleum); HM+P (soil contaminated by both).

in Bugang-Myeon, Sejong City, South Korea (Figure 1, S1); soils here exceeded the "third area concern" threshold for soil contamination under the Soil Environmental Conservation Act(Table S1), requiring remediation. The soils were located in a vadose zone above the water table (at a depth of 4 m). Three random samples were taken for HM+P and five for both HM and P, then mixed to single samples for each type that were analyzed three times. The average initial concentrations of Pb, Hg ,and total petroleum hydrocarbon (TPH) for each type are shown in Table 1. Remediation of all contaminated soils was conducted below the third area concern threshold for Pb (700 mgkg^{-1}), Hg (20 mgkg^{-1}), and TPH (2,000 mgkg^{-1}) and was evaluated for efficiency after five washing treatments.

Selection of washing agents and analysis

Soil washing agents were selected by pre-tests using H_2SO_4 and H_2O_2. The efficiency of the former at pH 1 was highest, matching the results of Lim (2005) and Paek et al. (2000). Thus, 0.2 M H_2SO_4 was used for washing HM; after diluting

35% H_2O_2 by 10%, it was also used for washing P; both agents were used simultaneously for HM+P. The analysis parameters were temperature, pH, moisture level, organic matter content, Pb, Hg, and TPH, assessed using standard methods of soil pollution analysis (Ministry of Environment, 2011)

Batch test

Both pH and the solid to liquid ratio (S/L; contaminated soil to diluted water) are important factors for calculating the whole volume during the washing process and to assess the effluent after washing (Zheng, et al., 2013). The washing agent must be selected according to the characteristics of the contaminated soil. The two agents (H_2SO_4 and H_2O_2) were tested by varying the pH range and S/L of each contaminated soil, then determining the optimal pH. Samples of 500 g each were prepared with particle size <0.2 mm using a 0.2 mm mesh. Jar tests were then conducted at pH values from 3–11 and S/L ratios of 1:2, 1:3, and 1:4. The optimal condition was considered to be reached when the remediation efficiency was highest.

Relocatable soil washing equipment

The relocatable soil washing equipment used in this study detaches and desorpts contaminated materials using physical mixing and impact effects based on changes in the rotation speed of the washing basin (Figure S1). After removing debris, the washing process separates particles larger than 25 mm using the hopper and vibration screen and sends the remaining contaminated soil to the washing tank containing washing agent. Subsequently, remediated soils < 0.2 mm are transferred to the sedimentation tank by sand pump while remediated soils> 0.2 mm are transferred to the dewatering screen and then discharged. Once the fine particles in the sedimentation tank settle, they are transferred to the sludge thickener and dewatering screen by a belt. Finally, the dewatering cake is discharged and dewatering water is recycled to the sedimentation tank.

RESULTS AND DISCUSSION

Optimal pH and S/L

For Pb in the combined HM+P soil (Figure 2), an S/L of 1:2 produced an average influent concentration of 174.9 mg kg^{-1} (157.6–194.6 mg kg^{-1}) and remediation efficiency of 86.9% (85.4–88.2%); the latter was highest at pH 5. An S/L of 1:3 produced a similar average influent concentration of 172.3 mg kg^{-1} (163.4–196.7 mg kg^{-1}) and remediation efficiency of 87.1% (85.2–87.7%); the latter was highest at pH 5 and 11. However, an S/L of 1:4 produced an average influent concentration of 159.8 mg kg^{-1} (136.7–183.4mg kg^{-1}) and remediation efficiency of 88.0% (86.2–89.7%); the latter was highest at pH 6 (Table S2). In other words, as the S/L changed from 1:2 to 1:4, the remediation efficiency increased slightly. The release of polluted materials from soil and increase in remediation efficiency at lower pH is similar to the results previous studies (Cheong et al., 1997; Lee et al., 2002; Lee et al., 2013; Kim, 2015).

For TPH in the combined HM+P soil (Figure 3), an S/L of 1:2 produced an average influent concentration of 205.9 mg kg^{-1} (119.3–338.2 mg kg^{-1}) and remediation efficiency of 86.7% (86.8–92.3%); the latter was highest at pH 5. An S/L of 1:3 produced an average influent concentration of 204.7 mg kg^{-1} (153.3–321.5 mg kg^{-1}) and remediation efficiency of 86.8% (79.2–90.1%); the latter was highest at pH 11. An S/L of 1:4 produced an average influent concentration of 185.0 mg kg^{-1} (119.1–314.8 mg kg^{-1}) and remediation efficiency of 88.0% (79.7–92.3%); the latter was highest at pH 5.

On average, Pb and TPH showed the highest remediation efficiency at pH 6 and 5, respectively. The remediation efficiency of Pb was similar at pH 5 and 6, while that of TPH was about 2% higher at pH 5 than pH 11, similar to previous studies which showed that remediation efficiency changes with S/L and is not higher for heavy metals (Choi, 2006, 2008). In other words, as the S/L changed from 1:2 to 1:4, the pattern of remediation efficiency was the same between Pb and TPH. Therefore, in this study, an S/L of 1:2 and pH 5 were selected as the optimal conditions.

On-site treatment of HM

Pb is an important factor for evaluating heavy metal pollution (Rötting et al., 2014; Ludajić et al., 2015). According to five random samples (Pb-1–5), the Pb concentration in the HM soil decreased most rapidly between the first and second washing (Table S3); after the second washing, this declined to 364.6 mg kg^{-1} with a remediation efficiency of 70.0%, satisfying the Pb standard threshold of 700 mg kg^{-1} (Figure 4a). However, the third washing reduced the average concentration to 119.6 mg kg^{-1} with an average remediation efficiency of 90.1%; the fourth and fifth washings did not change these values significantly, although the highest overall remediation efficiency (91.8%) was achieved after five washings. Therefore, the best performance for Pb in HM was achieved by three washings at an S/L of 1:2; a result similar to that reported by Kim (2013) for Pb removal from contaminated soil.

The behavior of Hg samples (Hg-1–5) was initially different from Pb in the HM soil, declining in concentration most rapidly after the first washing, but remaining mostly unchanged after the second washing (Figure 4b). The average concentration was 18.3 mg kg^{-1} after two washings, satisfying the Hg standard threshold of 20 mg kg^{-1}, with a

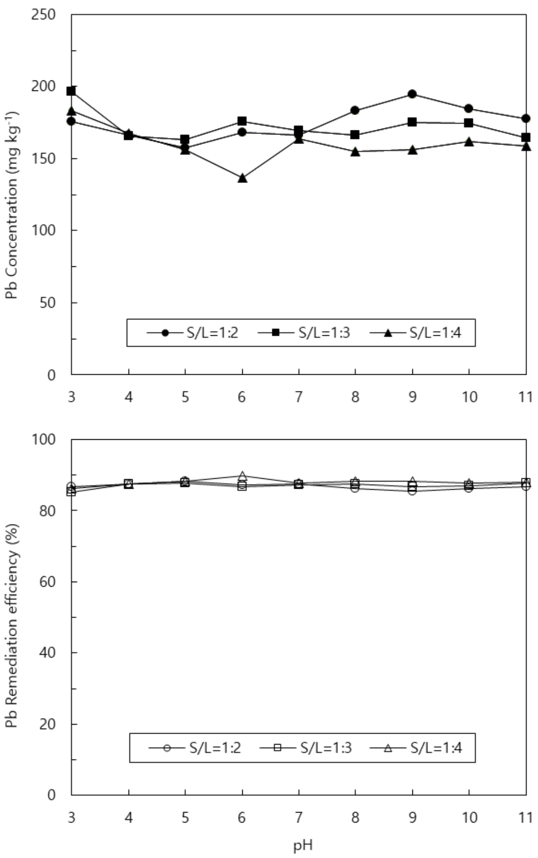

Figure 2: Variation of Pb concentration and remediation efficiency in the HM+P soil by pH for S/L ratios of 1:2, 1:3, and 1:4.

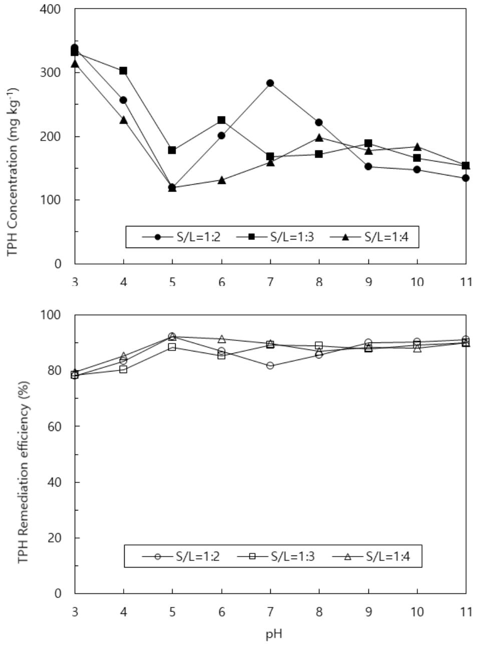

Figure 3: Variation of TPH concentration and remediation efficiency in the HM+P soil by pH for S/L ratios of 1:2, 1:3, and 1:4.

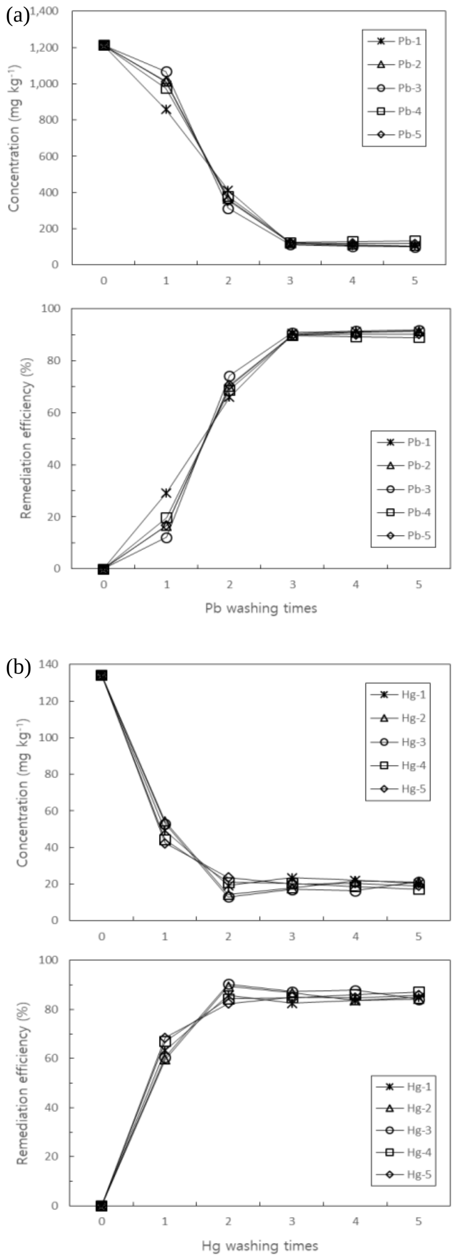

Figure 4: Variation of (a) Pb and (b) Hg concentrations and remediation efficiency by washing time in the HM soil.

remediation efficiency of 86.4%; neither changed significantly with subsequent washings. The highest remediation efficiency (90.3%) was achieved after two washings. Therefore, the best performance for Hg in HM was achieved by two washings at an S/L of 1:2.

On-site treatment of P

The TPH concentrations in five samples (TPH-1–5) in the P soil declined dramatically after the first washing but changed very little in subsequent washings. The remediation efficiency followed a similar pattern by increasing to 98.8% after the first washing with a slight increase in subsequent washings (Figure 5). This result is higher than the 90.0% efficiency reported by Lee et al. (2007) and Hwang et al. (2007) using H_2O_2. The first washing satisfies the TPH standard threshold of 2,000 mg kg^{-1}, although the highest remediation efficiency (99.1%) was achieved after five washings. Therefore, the best

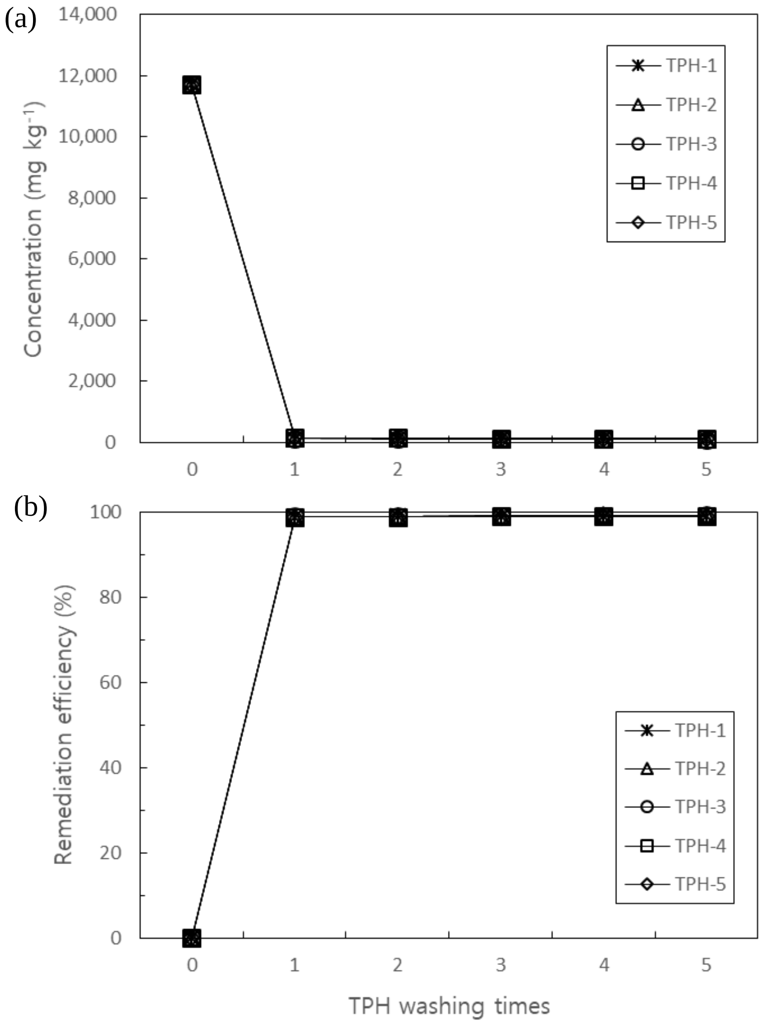

Figure 5: Variation of (a) TPH concentration and (b) remediation efficiency by washing time in the P soil.

performance for TPH in P was one washing at an S/L of 1:2, similar to the reports by Jang (2010) and Shin et al. (2014) of high TPH remediation efficiency for soil contaminated by petroleum after the first washing.

On-site treatment of HM+P

Three random samples of Pb (Pb-1–3), Hg (Hg-1–3), and TPH(TPH-1–3) were assessed in the combined HM+P soil. Pb followed a similar pattern as in the HM soil by declining rapidly through the first two washings and then changing very little (Figure 6a). The remediation efficiency similarly levelled out after two washings, although it peaked at 94.3% after five washings. The average Pb remediation efficiency of 91.0% was about 2–17% higher than that reported by Seol (2011) using HCl as a washing agent.

Hg concentration and remediation efficiency also followed similar trends to the HM soil, again changing rapidly after the first washing and only slightly after subsequent washings, satisfying the standard threshold (Figure 6b). The highest remediation efficiency (93.1%) occurred after the fifth washing. These results are similar to those reported by Kim (2015).

Similarly, TPH concentration and remediation followed similar trends to the P soil, with the first washing being by far the most effective (Figure 5c). The highest remediation efficiency (97.0%) occurred after two washings. These results are about 10% higher than that reported by Chun et al. (2007) with regard to TPH remediation efficiency in P soil after two washings using microorganism strains.

By comparing the washing agents, the remediation efficiency of Pb and/or Hg in HM+P was higher than in HM using 0.2M H_2SO_4 and P using 35% H_2O_2. The combined use of both washing agents was better than each alone, with an efficiency slightly higher by about 1% in HM+P. On the other hand, the sole use of 35% H_2O_2 produced a 3% higher remediation efficiency than use of both for TPH. The

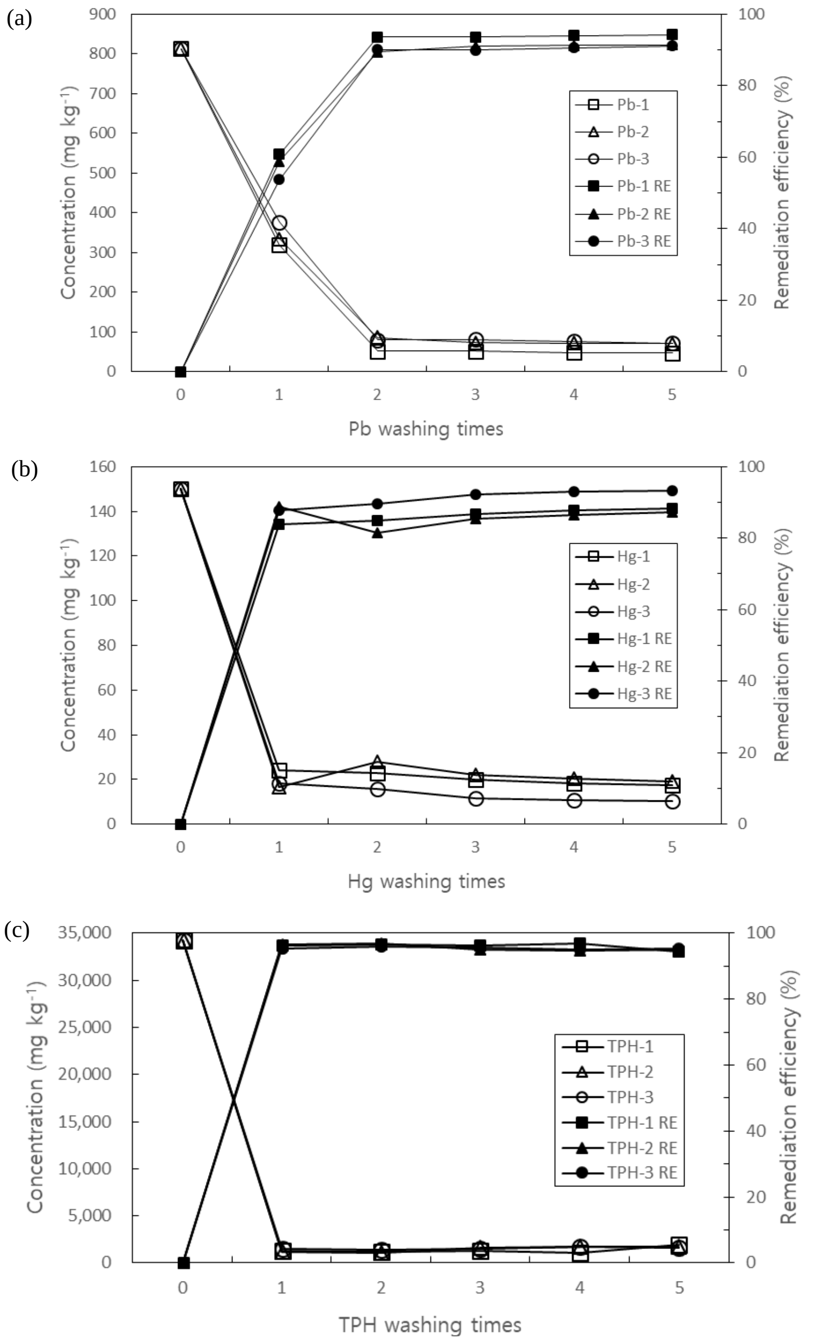

Figure 6: Variation of concentration and remediation efficiency for (a) Pb, (b) Hg, and (c) TPH by washing time in the HM+P soil; RE indicates remediation efficiency.

concentrations of Pb, Hg, and TPH were all reduced below the standard thresholds.

Conclusions

Preliminary tests on washing soil contaminated by heavy metals and petroleum using 0.2M H_2SO_4 and 35% H_2O_2 as washing agents with S/L ratios of 1:2, 1:3, and 1:4 and pH values from 3–11 showed that optimal conditions occurred at an S/L of 1:2 and a pH of 5. These conditions were then used to test remediation of Pb, Hg, and petroleum over five washing cycles using relocatable equipment. The remediation efficiency of soil contaminated only by the heavy metals Pb (90.1%) and Hg (86.4%) was best after the third and second washing using 0.2M H_2SO_4, respectively. Total petroleum hydrocarbon remediation efficiency in soil contaminated only by petroleum was highest (98.8%) after one washing using 35% H_2O_2. Soil contaminated with all three substances was stripped of Pb (by 91.0%), Hg (by 86.9%), and TPH (by 96.1%) after two, one, and one washings, respectively using both 0.2M H_2SO_4 and 35% H_2O_2. Therefore, the optimal approach to cleaning soil contaminated by heavy metals and petroleum under optimal conditions requires two washings for maximum results and efficiency.

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Supplementary Tables and Figure

Table S1: Thresholds for soil contamination in South Korea under the soil environmental conservation act (mgkg^{-1}).

Materials	First area		Second area		Third area	
	Concern	**Counter measure**	**Concern**	**Counter measure**	**Concern**	**Counter measure**
Cd	4	12	10	30	60	180
Cu	150	450	500	1,500	2,000	6,000
As	25	75	50	150	200	600
Hg	4	12	10	30	20	60
Pb	200	600	400	1,200	700	2,100
Cr^{+6}	5	15	15	45	40	120
Zn	300	900	600	1,800	2,000	5,000
Ni	100	300	200	600	500	1,500
F	400	800	400	800	800	2,000
Organophosphorus compound	10	-	10	-	30	-
PCBs	1	3	4	12	12	36
CN	2	5	2	5	120	300
Phenol	4	10	4	10	20	50
Benzene	1	3	1	3	3	9
Toluene	20	60	20	60	60	180
Ethylbenzene	50	150	50	150	340	1,020
Xylene	15	45	15	45	45	135
TPH	500	2,000	800	2,400	2,000	6,000
TCE	8	24	8	24	40	120
PCE	4	12	4	12	25	75
Benzopyrene	0.7	2	2	6	7	21

Table S2: Concentration and remediation efficiency (RE)of Pb and TPH by S/L ratio and pH in HM+P soil.

pH	S/L 1:2				S/L 1:3				S/L 1:4			
	Pb (mg kg^{-1})	**RE* (%)**	**TPH (mg kg^{-1})**	**RE* (%)**	**Pb (mg kg^{-1})**	**RE* (%)**	**TPH (mg kg^{-1})**	**RE* (%)**	**Pb (mg kg^{-1})**	**RE* (%)**	**TPH (mg kg^{-1})**	**RE* (%)**
3	175.5	86.8	338.2	78.1	196.7	85.2	321.5	79.2	183.4	86.2	314.8	79.7
4	166.4	87.5	256.3	83.4	165.8	87.6	299.9	80.6	167.2	87.5	226.4	85.4
5	157.6	88.2	119.3	92.3	163.4	87.7	173.7	88.8	155.9	88.3	119.1	92.3
6	168.4	87.4	201.0	87.0	175.6	86.8	201.1	87.0	136.7	89.7	131.7	91.5
7	166.0	87.5	283.4	81.7	169.4	87.3	167.5	89.2	163.7	87.7	159.5	89.7
8	183.2	86.3	221.5	85.7	166.0	87.5	171.6	88.9	154.9	88.4	198.6	87.2
9	194.6	85.4	151.8	90.2	174.9	86.9	188.1	87.8	155.9	88.3	177.5	88.5
10	184.6	86.2	147.4	90.5	174.6	86.9	166.0	89.3	161.7	87.9	183.2	88.2
11	177.4	86.7	134.6	91.3	164.1	87.7	153.3	90.1	158.9	88.1	154.3	90.0
AG*	174.9	86.9	205.9	86.7	172.3	87.1	204.7	86.8	159.8	88.0	185.0	88.0

*Initial concentrations of Pb and TPH were 1,333.0 mgkg^{-1}and 1,547.0 mgkg^{-1}, respectively; RE indicates remediation efficiency; AG indicates average values.

Table S3: Results and remediation efficiency of soil type by washing times.

				One washing		Two washings		Three washings		Four washings		Five washings	
Soil type	**Parameters**	**Initial Conc. (mgkg^{-1})**	**Samples**	**Conc. (mgkg^{-1})**	**Remediation efficiency (%)**	**Conc. (mgkg^{-1})**	**Remediation efficiency (%)**	**Conc. (mgkg^{-1})**	**Remediation efficiency (%)**	**Conc. (mgkg^{-1})**	**Remediation Efficiency (%)**	**Conc. (mgkg^{-1})**	**Remediation efficiency (%)**
HM	Pb	1,213.7	Pb-1	859.1	29.2	412.6	66.0	116.3	90.4	107.4	91.2	105.7	91.3
			Pb-2	1012.4	16.6	363.9	70.0	123.1	89.9	111.8	90.8	104.5	91.4
			Pb-3	1067.1	12.1	313.2	74.2	111.5	90.8	103.6	91.5	99.1	91.8
			Pb-4	974.6	19.7	377.9	68.9	124.6	89.7	130.2	89.3	133.0	89.0
			Pb-5	1010.9	16.7	355.3	70.7	122.5	89.9	120.1	90.1	118.3	90.3
			Average	984.8	18.9	364.6	70.0	119.6	90.1	114.6	90.6	112.1	90.8
	Hg	134.3	Hg-1	49.3	63.3	19.3	85.6	23.4	82.6	22.1	83.5	20.3	84.9
			Hg-2	54.3	59.6	14.3	89.4	17.8	86.7	21.8	83.8	21.1	84.3
			Hg-3	53.0	60.5	13.0	90.3	17.1	87.3	16.4	87.8	21.3	84.1
			Hg-4	44.2	67.1	21.0	84.4	20.5	84.7	18.7	86.1	17.3	87.1
			Hg-5	42.4	68.4	23.7	82.4	20.1	85.0	20.4	84.8	19.1	85.8
			Average	48.6	63.8	18.3	86.4	19.8	85.3	19.9	85.2	19.8	85.2
P	TPH	11,702.3	TPH-1	138.2	98.8	129.2	98.9	117.6	99.0	115.3	99.0	111.1	99.1
			TPH-2	136.4	98.8	131.7	98.9	107.3	99.1	103.9	99.1	101.7	99.1
			TPH-3	130.8	98.9	122.5	99.0	113.8	99.0	107.2	99.1	102.4	99.1
			TPH-4	146.3	98.7	149.2	98.7	133.7	98.9	130.1	98.9	129.6	98.9
			TPH-5	141.5	98.8	135.3	98.8	137.8	98.8	128.8	98.9	122.4	99.0
			Average	138.6	98.8	133.6	98.9	122.0	99.0	117.1	99.0	113.4	99.0
HM+P	Pb	813.3	Pb-1	318.5	60.8	52.3	93.6	51.5	93.7	48.4	94.0	46.7	94.3
			Pb-2	334.4	58.9	86.4	89.4	73.2	91.0	71.3	91.2	70.4	91.3
			Pb-3	376.0	53.8	80.5	90.1	81.4	90.0	76.8	90.6	71.9	91.2
			Average	343.0	57.8	73.1	91.0	68.7	91.6	65.5	91.9	63.0	92.3
	Hg	150.2	Hg-1	24.3	83.8	22.8	84.8	20.1	86.6	18.3	87.8	17.6	88.3
			Hg-2	16.7	88.9	27.8	81.5	22.0	85.4	20.3	86.5	19.2	87.2
			Hg-3	18.2	87.9	15.7	89.5	11.5	92.3	10.6	92.9	10.3	93.1
			Average	19.7	86.9	22.1	85.3	17.9	88.1	16.4	89.1	15.7	89.5
	TPH	34,189.4	TPH-1	1,292.1	96.2	1,162.3	96.6	1,263.5	96.3	1,048.7	96.9	1,897.7	94.4
			TPH-2	1,174.3	96.6	1,034.7	97.0	1,654.6	95.2	1,762.3	94.8	1,697.7	95.0
			TPH-3	1,545.5	95.5	1,354.4	96.0	1,475.2	95.7	1,662.9	95.1	1,598.4	95.3
			Average	1337.3	96.1	1183.8	96.5	1464.4	95.7	1491.3	95.6	1731.3	94.9

Acronyms: HM (soil contaminated solely by heavy metals); P (soil contaminated solely by petroleum); HM+P (soil contaminated by both).