Combined Use of Soil Washing and Incineration for Remediation of Dioxin Contaminated Soils in Southern Vietnam

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Introduction

Dioxin contamination in southern Vietnam is the result of the storage, loading, spillage, and handling of Agent Orange and other toxic herbicides during the US-Vietnam war. Dioxins are characterized by their low water solubility and their tendency to be strongly absorbed onto the surface of particulate matter, such as soils and sediments[1)]. Dioxins are resistant to degradation and present an ongoing toxicological threat to both human beings and wildlife. Therefore, full and cost-effective decontamination of dioxin contaminated soils is needed to give a complete solution to the problem. As the volume of contaminated soils is estimated very large, remediation of these sites requires applications of several technologies used in concert with each other, because no single technology is universally applicable. As shown in Figure 1, a combined use of soil washing and incineration appears to provide a cost-effective and environmentally proactive alternative to other remediation options. The principal use of soil washing is as a volume reduction technique in which the contaminants are concentrated into a smaller volume[2)]. Soil washing presents two key advantages of "*waste minimization*" and "*cost effectiveness*"[2)]. Lower remediation cost results from reduction of sheer volume of contaminated soils that must be treated by more expensive method. The contaminant concentrated soil (sludge cake) must be disposed of by further cleanup. Incineration is the most effective method to destroy chemically stable contaminants like dioxins[3), 4)]. However, incineration is also well-known to require a high energy demand and expensive cost[3), 4)]. It is considered that the combined use of soil washing and incineration makes it possible to achieve effective and cost-effective remediation of soils. The more the volume of dioxin contaminated soil which soil washing can accept and treat, the more cost-effective the combined use will be. Therefore, soil washing is needed to treat at the highest removal efficiency possible.

This paper provides an outline and key features of Shimizu's soil washing technology for organic and inorganic pollutants, and discusses the potential application of the combined use of soil washing and incineration to dioxin contaminated soils in southern Vietnam through a series of treatability tests.

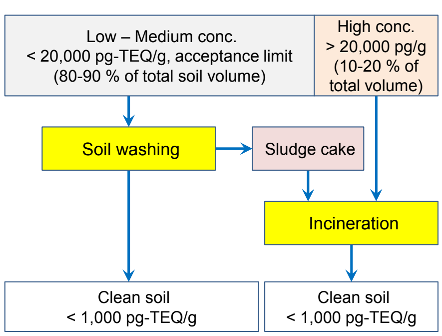

Figure 1: Combined use of soil washing and incineration

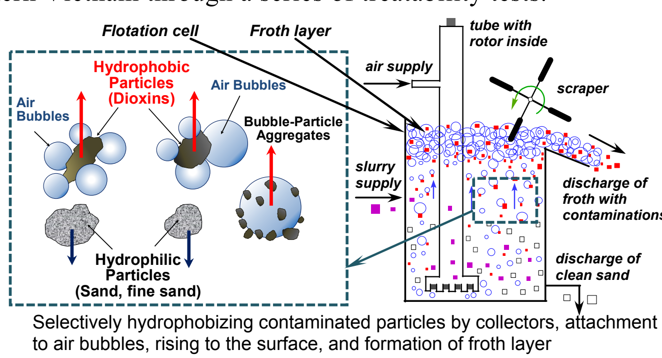

Figure 2: Contaminant removal in flotation process

Soil washing process and description

Shimizu's soil washing is a water-based process utilizing "physical separation" based on mining and mineral processing principles for removing a broad range of organic and inorganic contaminants from soils. The typical throughput range of commercial operation is 30-50 tons/hour. The operation units involved are: wet screen, hydrocyclone, attrition scrubber, froth flotation, up-flow column, spiral classifier, log washer, and so on. Hydrocyclone and flotation are the most important separation methods used in soil washing process. Hydrocyclone separates the fine soil from larger sand particles by the centrifugal force. Larger sand particles end up in the underflow; fine/light particles (silt, clay, organic) and water end up in the overflow.
In flotation, contaminant-bearing particles are conditioned such that they become selectively hydrophobic and non-contaminated particles hydrophilic. Adding a frothing agent and passing air bubbles through a slurry material removes the hydrophobic contaminated particles which attach themselves to the air bubbles and concentrate in a discharging surface froth as shown in Figure 2. Flotation has been successfully used to remove contaminants like heavy metals, herbicides, and dioxins from soils.

Materials and methods

A total of 9 soil samples were collected from selected "hot spots" in southern Vietnam. A bulk soil sample up to a depth of 0.5-1.5 m was collected by a mechanical excavator in a sampling point. 9 soil samples were composited and individual analyzed on TEQ content. Based on the analytical results, 5 soil samples were chosen to represent low (6,600pg-TEQ/g), average (12,000, 24,000pg-TEQ/g) and high (32,000, 57,000pg-TEQ/g) concentrations as shown in Table 1. PCDDs (2,3,7,8-TCDD) accounted for almost all of total TEQ in the soil samples.
A sequence of bench-scale treatability tests was performed to assess applicability of soil washing required to meet the cleanup target (1,000pg-TEQ/g) and to know volume reduction rates of feed soils. A sequence of treatability tests was carried out on batch operations of wet screen, hydrocyclone (d_{50} cut-point of 63μm), flotation, and sludge management (see Figure 4). Feed soil was screened to 2mm using a vibrating wet screen. The slurry underflow from the wet screening was then pumped to a hydrocyclone test unit. The underflow (sand) from the hydrocyclone unit was further processed through a flotation test device. The overflow and the froth, consisting of fine-grained material and water, were filtered and converted into sludge cake through sludge management.
Laboratory experiments were conducted to confirm the ability of incineration to destroy dioxin contaminants in sludge cake. Few studies have reported incineration performance on soils contaminated with very high levels of 2,3,7,8-TCDD[5)]. As shown in Figure 3, the laboratory test system consisted of an electric furnace and a set of air pollution control devices including a cooling water jacket, a filter, a cartridge, and adsorbing liquid for backup.

Table 1: Dioxin analysis and grain-size distribution

Untreated feed soils	Soil-A (pg-TEQ/g)	Soil-B (pg-TEQ/g)	Soil-C (pg-TEQ/g)	Soil-D (pg-TEQ/g)	Soil-E (pg-TEQ/g)	Sludge cake (composite) (pg-TEQ/g)
PCDDs	24,000	57,000	12,000	32,000	6,600	33,000
(2,3,7,8-TCDD)	(24,000)	(56,000)	(12,000)	(31,000)	(6,500)	(33,000)
PCDFs	100	190	49	110	24	180
Co-planar PCBs	1.5	1.6	0.23	0.69	0.25	1.5
Dioxins (total TEQ)	24,000	57,000	12,000	32,000	6,600	34,000
Dioxin criteria for soil	1,000 (pg-TEQ/g)					
Silt, clay (<63μm)	32.4%	24.9%	32.3%	25.5%	35.1%	Approx.100%
Sand (63μm-2mm)	51.7%	59.7%	52.3%	55.3%	50.2%	0.0%
Coarse (>2mm)	15.9%	15.4%	15.4%	19.2%	14.7%	0.0%
Sand + Coarse	67.6%	75.1%	67.7%	74.5%	64.9%	0.0%

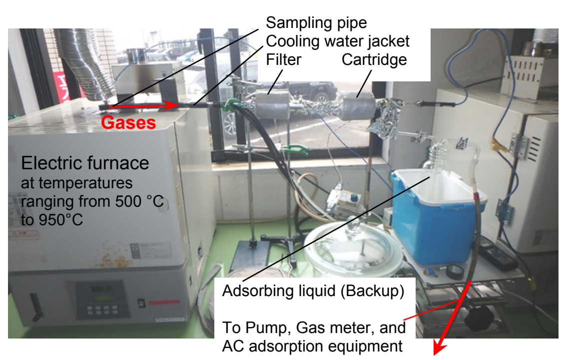

Figure 3: Laboratory test system of incineration

A composite sample (34,000pg-TEQ/g) of three sludge cakes of soil-A, soil-C, and soil-E was used as feed soil. In a series of experiments, 90g of feed soil was heated for 60min in the furnace at six different temperatures: 500°C, 650°C, 720°C, 800°C, 900°C, and 950°C. Mixed gas (N_2: 90%, O_2: 10%) was supplied into the furnace at a flow rate of 2L/min for 60min, and gases in the furnace were vacuumed by a pump at the same flow rate for 60min. As feed soil heated up, the contaminants volatilized, changed into gases and most were destroyed. Nearly all of dioxin compounds (particulate and gaseous phase) in gases and condensed water were captured through the air pollution control devices. After each experiment, the soil and gas (particulate and gaseous phase) samples were collected and analyzed for total dioxin/TEQ contents or concentrations.

Results and discussion

As shown in Table 1, the grain-size distributions of five samples learn that the sand fraction (63μm-2mm) ranged from 50% to 60%, and that coarse fraction (>2mm) varied from 15% to 19%. The silt/clay fraction (<63μm) was between 25% and 35%. As the sum of sand and coarse fractions represents approximate percentages of clean product recoveries possible, the recovery rare of clean products (sand plus gravel) was expected to be between 65% and 75%. Table 2 shows soil washing performance on 5 soil samples. Total TEQ contents of clean sands ranged from 200 to 3,200pg/g as compared to the feed soils of 6,600 to 57,000pg/g. The removal efficiencies were between 93.6% and 97.6%, and their average was 95.6%. It is well known that dioxin contaminants tend to bind, either chemically and physically, to organic, clay, and silt particles, and that these smaller particles tend to be, loosely or tightly, attached to larger sand particles by adhesion and compaction. Scrubbing at the flotation stage can effectively detach and liberate smaller contaminated particles from larger sand particles. Figure 4 shows the process route of soil-A treatment. The total TEQ contents were 9,200pg/g for the cyclone underflow, 580pg/g

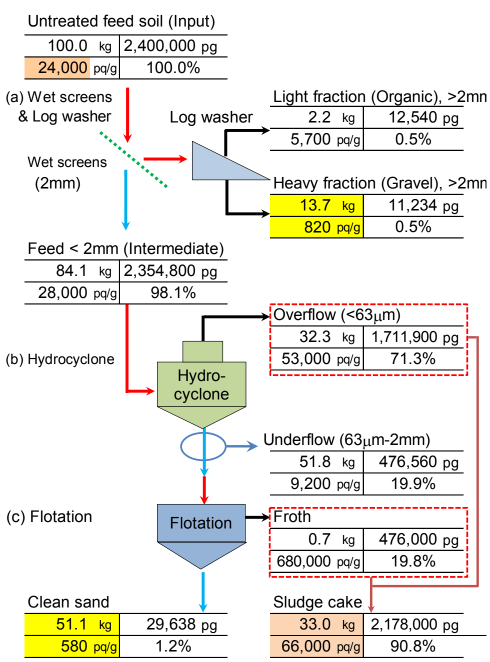

Figure 4: Process route of soil-A treatment

Table 2: Soil washing performance on dioxin contaminated soil

	Untreated feed soil (input)	*Hydrocyclone*		*Flotation*			Removal efficiency	Sludge cake (Overflow plus Froth)
		Underflow (intermediate)	Overflow (output)	Clean sand (output)		Froth (output)		
	(pg-TEQ/g)	(pg-TEQ/g)	(pg-TEQ/g)	(pg-TEQ/g)	*Criteria*	(pg-TEQ/g)		(pg-TEQ/g)
Soil-A	24,000	9,200	53,000	580	*Yes*	680,000	97.6%	66,000
Soil-B	57,000	11,000	160,000	3,200	*No*	460,000	94.4%	170,000
Soil-C	12,000	1,900	25,000	770	*Yes*	53,000	93.6%	26,000
Soil-D	32,000	5,700	66,000	1,500	*No*	300,000	95.3%	73,000
Soil-E	6,600	970	18,000	200	*Yes*	17,000	97.0%	18,000
						Average	95.6%	

Table 3: Total dioxin/TEQ contents in treated soil samples

	Feed soil (Sludge cake)	Treated soil					
		500°C	650°C	720°C	800°C (1st)	800°C (2nd)	900°C
Total dioxin content (pg/g)	61,000	0.3	0.2	N.D.	N.D.	N.D.	5.6
Total TEQ content (pg/g)	34,000	0	0	0	0	0	0.0017

Remarks
- Effective inner volume of furnace: 9.6 L
- Incineration temperature: 500°C, 650°C, 720°C, 800°C, and 900°C
- Residence time of feed soil: 60 min
- Supply of mixed gas (N_2: 90%, O_2: 10%) into furnace: 2 L/min, 60min
- Gas sampling (vacuuming): 2 L/min, 60min
- Residence time of gas: 4.8 min

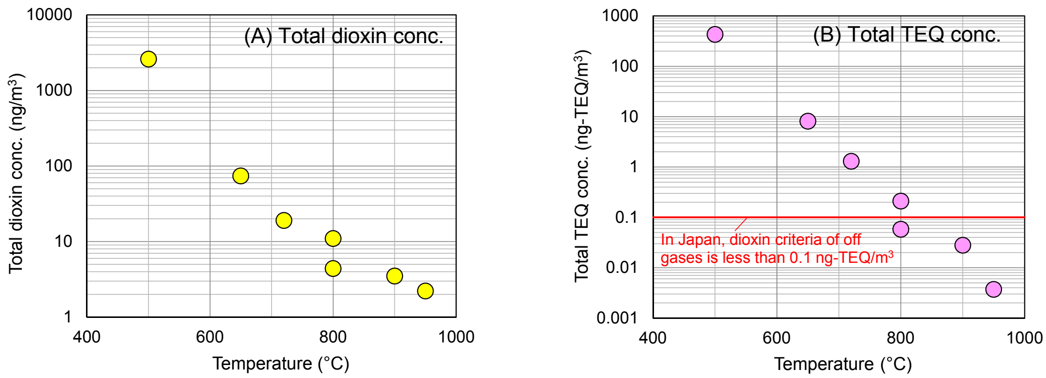

Figure 5: Total dioxin/TEQ concentrations of the gas samples in the temperature range 500 to 950°C

for the clean sand respectively. The removal efficiency of hydrocyclone was 61.7%, and it was greatly improved to 97.6%, when the hydrocyclone system was combined with the flotation system. The above results indicate that the soil washing system with flotation makes it possible to treat soils contaminated with up to 20,000pg-TEQ/g, and to produce clean sands below the dioxin criteria (1,000pg-TEQ/g).

Table 3 shows incineration performance on the sludge cake sample of 34,000pg-TEQ/g in the temperature range 500 to 900°C. After heating at 500°C or higher for 60min, the total TEQ contents of the treated samples were 0pg-TEQ/g or near-zero and the removal ratios were more than 99.99%. This result indicates that incineration at 500°C or higher for 60min can completely remove total TEQ of dioxins from sludge cake. Total dioxin/TEQ concentrations of the gas samples (particulate and gaseous phase) are plotted in Figure 5. The residence time of gases in the furnace was about 4.8min. Total dioxin concentration means the sum of PCDDs, PCDFs, and Co-Planar PCBs concentrations. Figure 5(A) shows that as the temperature was raised, the total dioxin concentration declined significantly. This suggests that the higher the temperature, the more and the faster the thermal decomposition of dioxin compounds. In Japan, for large incineration plants the dioxin criteria of off gases is less than 0.1ng-TEQ/m^3. Figure 5(B) shows that total TEQ concentrations were below 0.1ng-TEQ/m^3 in the temperature range above 800°C without using a secondary furnace for further heating and destruction.

The above test results with soil samples collected from hot spots confirmed two important factors to applying the combined use; 1) waste minimization by soil washing at a rather high removal efficiency of 93.6-97.6%, and 2) almost complete decomposition of dioxin compounds by incineration in the temperatures range 800 to 900°C. The combined use is expected to be practically used in southern Vietnam for cost-effective soil remediation.

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