

REMEDIATION OF DIOXIN-CONTAMINATED SOIL BY SUCCESSIVE ETHANOL WASHING

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Introduction

Although soil highly contaminated with dioxins was found in Nosecho, Japan, in April 1998, being caused by a municipal incinerator, remediation has yet to commence due to protests by the local citizenry. Unlike incineration treatment of dioxin-contaminated soil, a method having received considerable research attention recently, removal of dioxins by ethanol washing is accomplished without heating, *i.e.*, at room temperature and atmospheric pressure. Moreover, such treatment is relatively simple and does not require large components/equipment which make it highly suitable for application using a mobile treatment facility; a concept that is considered to more likely gain public acceptance and promote faster remediation of areas having soil suffering from dioxin contamination.

With this factor in mind, we have been developing a multi-stage ethanol washing process. Here, we report on the suitability of applying this treatment method to actual dioxin-contaminated soil found in Nosecho, Japan. The removal mechanism is also discussed.

Method

Analysis of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) was performed using gas chromatography high-resolution mass spectrometry (HRGC-MS; GC, Hewlett Packard, 5890-2; MS, Nihondenshi, Mstation-700).¹ Calculations were based on the International Toxicity Equivalency Factor determined by WHO/IPCS in 1998. The concentration of coplanar polychlorinated biphenyls (co-PCBs) was not determined.

Soil consisted of light-colored particles having the following properties: mesh size, No. 10–20; pH, 6.5; water content, 2.5%; and organic content, 1.6%.²

All washing was done at 30°C. Using 6-g samples, 18 mL of fresh ethanol was added and the mixture washed rotationally (200 rpm) for 24 h. Then, after removing 12 mL of supernatant and adding 12 mL of fresh ethanol, the soil was similarly washed again for a total of seven washings. Figure 1 shows a diagram of the employed ethanol washing process. All analyses were performed in triplicate ($n = 3$), with average values being indicated in results.

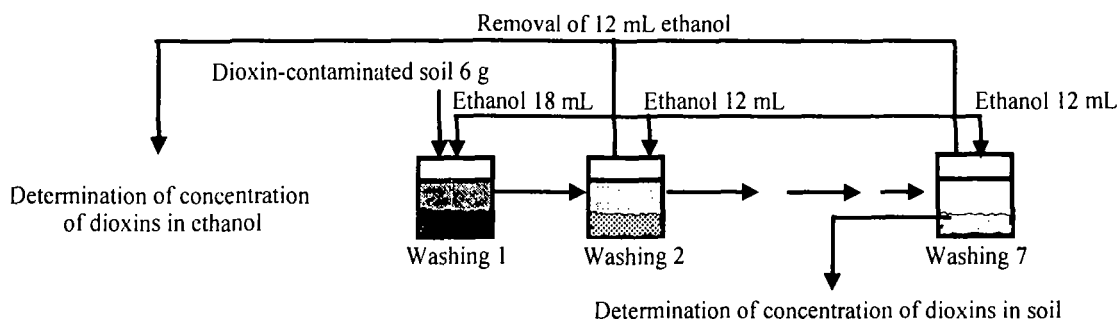


Fig. 1 Diagram of employed ethanol washing treatment method.

Results and discussion

In previous work, we demonstrated that ethanol is the best washing solvent for soil contaminated with polycyclic aromatic hydrocarbons (PAHs).³ It is generally known that dioxins and PAHs exhibit similar chemical properties and environmental behaviors. We also used isopropyl alcohol (IPA) as a washing solvent because it has been frequently used as a hydrogen donating solvent for ultraviolet degradation of PCBs.⁴

Figure 2 shows dioxin concentrations after each ethanol or IPA washing, i.e., regression curves of $\ln C_t/C_0$ in which C_0 is the initial concentration in soil and C_t the concentration in soil after t washings. Note that for ethanol, a pseudo first-order reaction appears in relation to the number of times washed for 2378-T4CDD, 12378-P5CDD, and 23478-P5CDF, each of which shows strong toxicity in comparison with other 2378-substituted isomers, i.e., a TEQ higher than 0.5. Also, different slopes which are constant first order are present between washing 1 and 3 or 4 and 7 because the affect of washing successively decreases due to strong dioxin adsorption by the soil matrix.

These results indicate that removal efficiency for each washing is proportional to the remaining dioxin concentration in the soil. In contrast to ethanol, with the exception of 234678-H6CDF, removal of dioxins by IPA does not follow a first-order effect.

Table 1 summarizes the concentrations of 2378-substituted dioxins before and after multiple washings with ethanol or IPA, where after 7 washings the dioxin concentration is about 450 or 1279 pg-TEQ/kg, respectively, and for all 2378-substituted isomers, more than 90 or 78% are removed, respectively.

Clearly then, ethanol is more suitable as a washing solvent for actual dioxin-contaminated soil in that after multi-stage ethanol or IPA washings the dioxin removal efficiency was about 95.1 or 85.9%, respectively. These results demonstrate that the proposed ethanol washing treatment can be effectively applied to remove dioxins from actual soils.

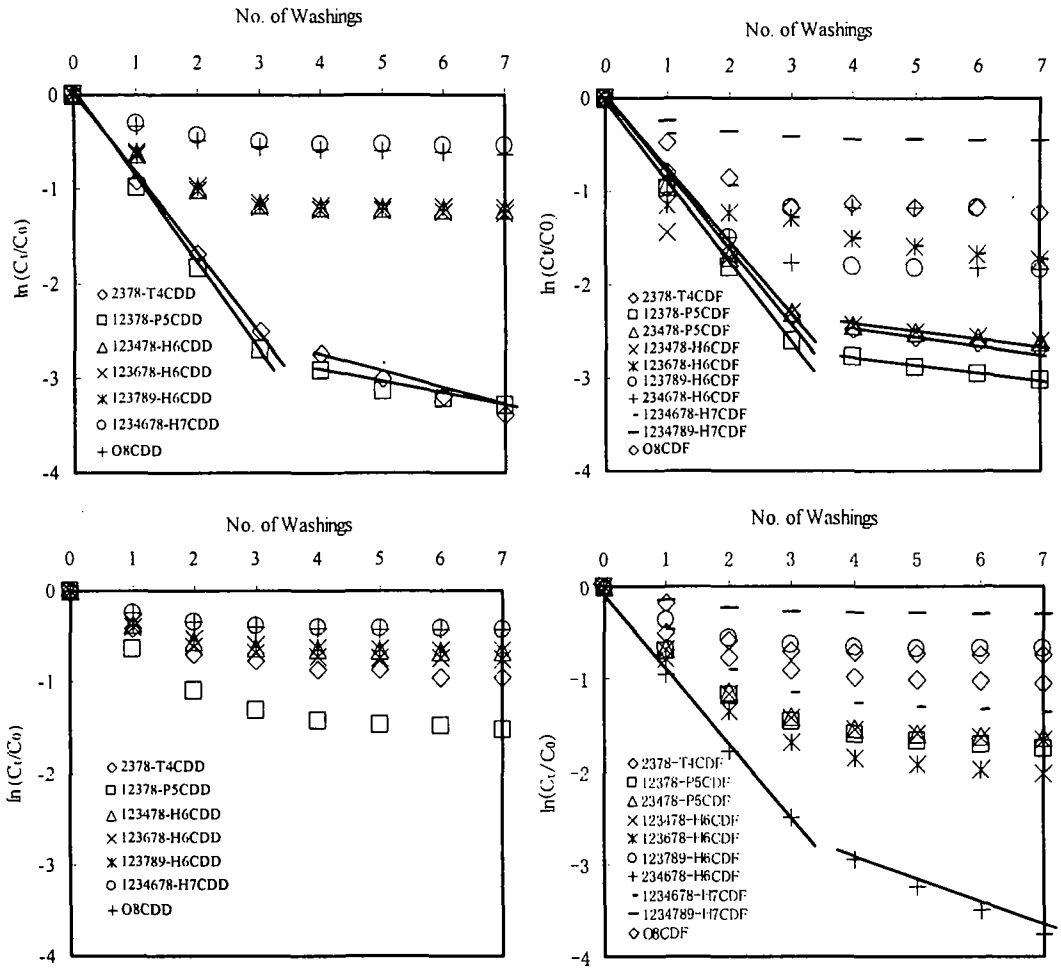


Fig. 2 Comparison of the washing effect using ethanol (upper) or IPA (lower) to remove 2378-substituted dioxins from actual soil.

Table 1 2378-substituted isomer concentrations in actual soil before and after washing seven times with ethanol or IPA.

Dioxin	Initial (pg-TEQ/g)	Final (pg-TEQ/g)		Removal Efficiency (%)	
		Ethanol	IPA	Ethanol	IPA
2378-T4CDD	31.6	3.3	7.0	89.5	77.9
12378-P5CDD	678.9	49.1	141.3	92.8	79.2
123478-H6CDD	320.7	10.8	33.4	96.6	89.6
123678-H6CDD	390.3	14.1	42.5	96.4	89.1
123789-H6CDD	325.7	12.8	37.5	96.1	88.5
1234678-H7CDD	217.2	9.7	11.9	95.5	94.5
O8CDD	3.6	0.2	0.2	95.1	94.2
2378-T4CDF	35.0	2.5	6.6	92.9	81.3
12378-P5CDF	74.9	4.3	13.8	94.3	81.6
23478-P5CDF	1990.6	114.1	315.2	94.3	84.2
123478-H6CDF	679.1	29.6	104.4	95.6	84.6
123678-H6CDF	859.2	35.6	135.9	95.9	84.2
123789-H6CDF	1325.9	25.5	85.7	98.1	93.5
234678-H6CDF	1778.5	98.2	305.9	94.5	82.8
1234678-H7CDF	228.7	33.2	31.9	85.5	86.0
1234789-H7CDF	157.4	6.6	5.7	95.8	96.4
O8CDF	4.7	0.4	0.4	92.2	90.9
Total PCDDs/DFs	9102.0	450.0	1279.1	95.1	85.9

References

1. Waste Management Division, Water Supply and Environmental Sanitation Department, Environmental Health Bureau of the Ministry of Health and Welfare (1997) Standard manual for determination and analysis of dioxins in waste treatment process. Environmental Health Bureau of the Ministry of Health and Welfare (in Japanese).
2. Japan Soil Fertilizer Science Association (1992) Standard Methods for the Examination of Soil, p. 21 (in Japanese).
3. Lee B.-D. and Hosomi M. (2000) Ethanol washing of PAH-contaminated soil and Fenton oxidation of washing solution, *J. Mater. Cycles Waste Manag.* 2(1), 24–30.
4. Toru Nishiwaki, Atsuyuki Ninomiya, Shizuo Yamanaka and Kinzi Anda (1972) *Nihonkagakugakkaishi*, 11, 2225 (in Japanese).