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Distribution of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the landfill site for solidified monoliths of fly ash

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Abstract

One landfill site, which co-treated solidified monoliths of fly ash and bottom ash, was investigated comprehensively to characterize its PCDD/F distribution. The solidified monoliths, soil, banyan leaves, groundwater in the monitoring wells and the treated landfill leachates in this landfill site for solidified monoliths of fly ash were all sampled to clarify their PCDD/F characteristics. Although the PCDD/F leaching concentrations were extremely lower than the Taiwan PCDD/F TCLP regulation of solidified monoliths, the PCDD/F contents in the surface soils of the landfill site are 460 times higher than that of urban soils and the highest value is 2.8 times higher than the Taiwan soil regulation (1000 ng I-TEQ kg^{-1}). The elevated PCDD/F contents in the soil reveal their potential for causing adverse health risk for humans, including the pathway of resuspension of soil particles and volatilization of PCDD/Fs from soil. The PCDD/F concentrations in the groundwater and the treated landfill leachates of the landfill site for solidified monoliths were both higher than that in the control samples, suggesting its potential to be a PCDD/F source of nearby water environment. Without proper control and management, landfill sites for solidified monoliths of fly ash can seriously hazard the surrounding environment, therefore, are important to consider.

Keywords: Dioxin; Landfill site; Solidified monoliths; Soil; Groundwater

1. Introduction

After polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were discovered in the flue gases and fly ash of municipal solid waste incinerators (MSWIs) in 1977 [1], PCDD/Fs have become a serious issue in many countries, because of their toxicological effects and associated adverse health implications.

After upgrading of air pollution control devices (APCDs) and using activated carbon for removing PCDD/Fs, the PCDD/F emission from MSWIs could keep the stringent emission limit (0.1 ng I-TEQ Nm^{-3}). However, PCDD/Fs are merely transferred from flue gas to fly ash after the treatments of APCDs and people's concern regarding the problem of PCDD/F has also shifted to the final treatment of fly ash.

In Taiwan, the fly ash generated from MSWIs must be solidified by cement and then evaluated by the toxicity characterization leaching procedure (TCLP) to forward to the landfill sites. The ratio of cement, additives and water to fly ash is not regulated but is usually about 0.2, 0.02 and 0.3, respectively. The TCLP regulation of solidified monoliths of fly ash for PCDD/F leaching concentrations is 0.001 mg I-TEQ L^{-1}. In Japan, however, leaching regulation for PCDD/Fs has not yet been established, although 3000 ng I-TEQ kg^{-1} has been designated as a content regulation for landfill, which was determined by reference to the environmental soil standard (1000 ng I-TEQ kg^{-1}) set up in terms of the risk posed by direct oral intake of soil particles containing PCDD/Fs [2].

Many studies indicate that the PCDD/Fs (particularly the more highly chlorinated congeners) possess such strong hydrophobicity and exhibit little downward mobility once deposited in or on soil [2–6]. Orazio et al. [3] investigated the soil in the vicinity of a site contaminated by PCDD/Fs. The sandy loam soil, with a pH of 8.1, contained 1.7% organic matter, 44% sand, 34.5% silt and 21.2% clay. Over a 15-month period, measurable degradation was found to occur only in the case of di- and trichloro congeners. The degradation of tetra through octachloro congeners was minimal. Cerlesi et al. [6] estimated

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a soil half-life for TCDD of approximately 9.1 year based on the field measurements and an exponential degradation model. Hagenmaier et al. [4] reported that only little movement and neither an appreciable loss of PCDD and PCDF nor significant changes in the homologue and isomer patterns could be detected in the contaminated soil samples taken in 1981, 1987 and 1989 from the same depth, which were in the surroundings of two industrial plants.

However, Osako et al. [7] found that the coexistence of dissolved organic materials (DOMs), which originating from bottom ash mixed and buried with the fly ash, could enhance the leachability of PCDD/Fs, thus contributing to the vertical movement and leaching behavior of PCDD/Fs in the landfill layers. Osako and Kim [2] also found that the enhancement of PCDD/F leachability by sufficient addition of surface active agents (SAAs) in the leaching test, which was assumed that SAAs are from municipal solid waste just like DOMs from the bottom ash, whereas addition of a smaller quantity depressed the leachability. The higher the degree of chlorination in PCDD/F homologues, the more effectively the SAAs enhanced the leachability. Schramm et al. [8] conducted the leaching experiments with fly ash and soil by fire-extinguishing water and found the significant amounts of PCDD/F, especially highly chlorinated congeners in the leachate.

There are 19 MSWIs in Taiwan, which currently produce approximately 168,000 tons of fly ash per year. After solidification, the solidified monoliths of fly ash are sent to the 15 landfill sites, which treat not only solidified monoliths but also the municipal waste or bottom ash. The solidification treatments of fly ash in Taiwan are all base on the cement solidification and can be further divided into two categories roughly according to the additives, that is, phosphate compound and sulfur compound [9]. In this study, one landfill site, which co-treated solidified monoliths of fly ash and bottom ash, was investigated comprehensively to characterize its PCDD/F distribution. Although only one landfill site for solidified monoliths of fly ash is chosen, however, the similar solidification treatments of fly ash and landfill process should make the obtained results possess representative of other landfill sites. The solidified monoliths of fly ash, soil, banyan leaves, groundwater in the monitoring wells and the treated landfill leachates were sampled and measured for PCDD/Fs to characterize the PCDD/F distribution in the landfill site for solidified monoliths of fly ash.

2. Experimental

2.1. Solidified monoliths of fly ash

Six solidified monolith samples, which come from two MSWIs, were sampled from the temporary storage area of the landfill site and conduct by TCLP to measure the PCDD/F leaching concentrations. Another six solidified monoliths samples were sampled for PCDD/F content. All the solidified monolith samplings followed the Taiwan standard sampling method for industrial waste (NIEA R118.01B) for obtaining the representative samples.

For the leaching test, 100 g of solidified monolith sample was extracted by batch leaching test in accordance to the Taiwan standard leaching test (NIEA R201.13C), while 1 g of solidified monolith sample was used for measuring PCDD/F content.

2.2. Soil

Within an area of approximately 36 m^2, nine points were chosen and each point collected about 500 g of soil. All these soils are combined together and considered as one representative sample, following the Taiwan standard sampling measure for soil (NIEA S102.60B).

Three representative surface soil samples (at depths from 0 to 5 cm) and another three inner soil samples (the same places but at depths from 5 to 15 cm) were collected from the landfill site. For comparison, another six soil samples were collected from urban areas about 10 km far from landfill sites.

After naturally weathering to dry in room, 10 g soil was used for analytical purposes.

2.3. Banyan leaves

Because banyan is the most common plant in Taiwan, we choose banyan leaves as indicator for easily comparing with that of other places in Taiwan. Six banyan leaf samples were collected from the landfill site, while another six leaf samples were collected from the same places with urban soil.

Each leaf sample was collected from 3–5 banyans and each banyan was sampled about 500 g of leaf by cutting at a height of approximately 1–2 m from the ground. All these leaf were combined together and immediately packed in aluminum foils and considered as a representative sample. Subsequently, samples were naturally weathered to dry in room and kept until analysis. After pretreatment by grinding them with a cutting mill to a fine powder, 20 g were used for analytical purposes.

2.4. Groundwater and the treated landfill leachates

Three groundwater samples were sampled from the upstream, midstream and downstream monitoring wells of the landfill site, respectively and followed the Taiwan standard method of groundwater sampling (NIEA W103.52B). Another one groundwater and four river water samples sampled from two different rivers, which collected from other county were considered as the background samples. Because PCDD/Fs hardly dissolve in water, the sampled water quantity must be adequate for PCDD/F analysis. Samples of water were collected in accordance with our patent “The device for sampling micro organic pollutant from water” (Taiwan patent no. 00547131). The device was equipped with a quartz-fiber filter for sampling particle-phase PCDD/Fs, and followed by a cartridge (polyurethane foam, PUF) for sampling liquid-phase PCDD/Fs, respectively. The sampling flow rate was about 0.4–0.6 L min^{-1} to finally reach water sample of 1000 L. Similarly, three samples of the treated landfill leachates were sampled from leachate treatment plant.

2.5. Analyses of PCDD/Fs

PCDD/F analyses of fly ash solidification, soil, water and banyan leaves samples followed the Taiwan PCDD/F analysis method NIEA M801.10B. All chemical analyses were carried out by the Super Micro Mass Research and Technology Center in Cheng Shiu University—the accredited laboratory for analyses of PCDD/F and now the only one accredited laboratory for PCDD/F analysis of soil. The sample analyses were performed according to the standard procedures. Two high-resolution gas chomatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/Fs analyses (one for analyzing solidified monoliths and soil samples, and the other for water and leaf samples). The HRGC (Hewlett Packard 6970 Series gas, CA, USA) was equipped with a DB-5MS fused silica capillary column ($L = 60$ m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA, USA), and with a splitless injection. Helium was used as the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used with resolving power at 10,000. The electron energy and source temperature were specified at 35 eV and 250 °C, respectively. Details are given in Wang et al. [10].

3. Results and discussion

3.1. PCDD/F leaching concentrations and PCDD/F contents of the solidified monoliths

The mean PCDD/F leaching concentration of the solidified monoliths of the fly ash was listed in Table 1 and was 0.0776 pg I-TEQ L^{-1} (R.S.D.: 133%). Comparing to the Taiwan PCDD/F TCLP regulation of solidified monoliths (0.001 mg I-TEQ L^{-1}), the PCDD/F leaching concentration is 7.8 E-8 times lower than the regulation.

The mean PCDD/F content in the solidified monoliths of the fly ash was listed in Table 2 and was 367 ng I-TEQ kg^{-1} (Range: 134–561 ng I-TEQ kg^{-1}, R.S.D.: 41.0%), which was about one tenth of the Japanese content standard for landfill (3000 ng I-TEQ kg^{-1}). No matter which regulation is, these solidified monoliths of the fly ash were qualified to be sent to the landfill site.

Table 1
PCDD/F leaching concentrations of the solidified monoliths of the fly ash

PCDD/Fs	Range ($n = 6$)	Mean ($n = 6$)	R.S.D. (%)
PCDDs (pg L^{-1})	0.260–34.1	6.96	193
PCDFs (pg L^{-1})	0.0827–9.08	2.17	160
PCDDs/PCDFs ratio	1.00–3.75	2.32	46
Total PCDD/Fs (pg L^{-1})	0.343–43.1	9.13	185
Total I-TEQ (pg I-TEQ L^{-1})	0.00243–0.247	0.0776	133

Table 2
PCDD/F content in the solidified monoliths of fly ash

PCDD/Fs	Range ($n = 6$)	Mean ($n = 6$)	R.S.D. (%)
PCDDs (ng kg^{-1})	1100–3630	2560	32.7
PCDFs (ng kg^{-1})	899–4120	2800	41.7
PCDDs/PCDFs ratio	0.803–1.23	0.970	18.1
Total PCDD/Fs (ng kg^{-1})	2000–7750	5360	37.2
Total I-TEQ (ng I-TEQ kg^{-1})	134–561	367	41.0

3.2. PCDD/F contents in the soil

The PCDD/F contents in urban soils and the surface and inner soils of the landfill site were both listed in Table 3. The mean PCDD/F content in the urban soils was 2.74 ng I-TEQ kg^{-1} (R.S.D.: 110%). On the basis of various published studies, US EPA [11] estimated that the mean TEQ values for background urban and rural soils in United States are 13.4 and 4.10 ng I-TEQ kg^{-1}, respectively and are comparable to that obtained in our study.

The mean PCDD/F contents in surface soils and inner soils of the landfill site were 1260 ng I-TEQ kg^{-1} (92.4–2810 ng I-TEQ kg^{-1}, R.S.D.: 111%) and 437 ng I-TEQ kg^{-1} (200–667 ng I-TEQ kg^{-1}, 53.5%), respectively, which were 3.4 (=1260/367) and 1.2 (=437/367) times higher than that in the solidified monoliths of the fly ash, respectively. It reveals that the soils had been contaminated by PCDD/Fs and the polluted region had been downward to inner soils. Because the organic matter of the soil, the released or leaching PCDD/Fs from solidified monoliths had finally accumulated in soils to reach higher PCDD/F content, even higher than that of the solidified monoliths.

Comparison with urban soils, the PCDD/F contents in the surface soils of the landfill site are 460 (=1260/2.74) times higher and the highest value obtained in this study is 2.8 times higher than the Taiwan soil regulation (1000 ng I-TEQ kg^{-1}), suggesting their potential for causing adverse health risk for humans, including the pathway of resuspension of soil particles and volatilization of PCDD/Fs from soil. Lee et al. [12] quantified PCBs in air samples taken every 6 h over a 7-day period in the summer at a rural site in England and found a strong correlation between air temperature and PCB congener concentrations. In the United Kingdom, Duarte-Davidson et al. [13] measured PCDD/F concentrations in various environmental compartments and suggested that volatilization from soils was one of the major sources of PCDD/Fs to the atmosphere.

Fig. 1 shows the congener profiles of the seventeen 2,3,7,8 chlorinated substituted PCDD/Fs (mean ± S.D.) detected from the solidified monoliths and soil samples. Each selected congener was normalized by reference to the total weight of all 2,3,7,8-congeners. We found that the PCDD/F congener profiles of the solidified monoliths and soils of the landfill site were similar, which dominated by 1,2,3,4,6,7,8-HpCDD, OCDD, 1,2,3,4,6,7,8-HpCDF and OCDF, and different to that of urban soils, which dominated by only OCDD. It confirms that the soils of the landfill site, which are 460 times higher than that of urban

Table 3
PCDD/F content in the soil

PCDD/Fs	Urban soil		Landfill site			
	Surface soil ($n=3$)		Surface soil ($n=3$)		Inner soil ($n=3$)	
	Mean	R.S.D. (%)	Mean	R.S.D. (%)	Mean	R.S.D. (%)
PCDD (ng kg^{-1})	129	72.0	11000	90.2	5360	37.0
PCDF (ng kg^{-1})	21.7	105	12300	118	3920	49.9
PCDDs/PCDFs ratio	18.6	142	1.51	63.4	1.46	21.9
Total PCDD/Fs (ng kg^{-1})	151	54.9	23400	105	9280	42.2
Total I-TEQ (ng I-TEQ kg^{-1})	2.74	110	1260	111	437	53.5

soils and the highest value is 2.8 times higher than the Taiwan soil regulation (1000 ng I-TEQ kg^{-1}), are contaminated by the solidified monoliths of fly ash.

3.3. PCDD/F contents in the banyan leaves

The mean PCDD/F content in the banyan leaves of urban area and the landfill site were listed in Table 4 and were 2.48 ng I-TEQ kg^{-1} (1.29–3.82 ng I-TEQ kg^{-1}, 42.6%) and 4.20 ng I-TEQ kg^{-1} (1.92–11.4 ng I-TEQ kg^{-1}, 85.5%), respectively. The *t*-test shows that there is no statistical significant difference between these two data sets. However, one of the banyan leaf samples possesses high PCDD/F content, 11.4 ng I-TEQ kg^{-1} and it may be result from the particulate deposition from ambient air or soil with elevated PCDD/F content.

3.4. PCDD/F concentrations in groundwater and the treated landfill leachates

Table 5 lists the PCDD/F concentrations in the control group (one groundwater and four river water samples), the groundwater of monitoring wells and the treated landfill leachates,

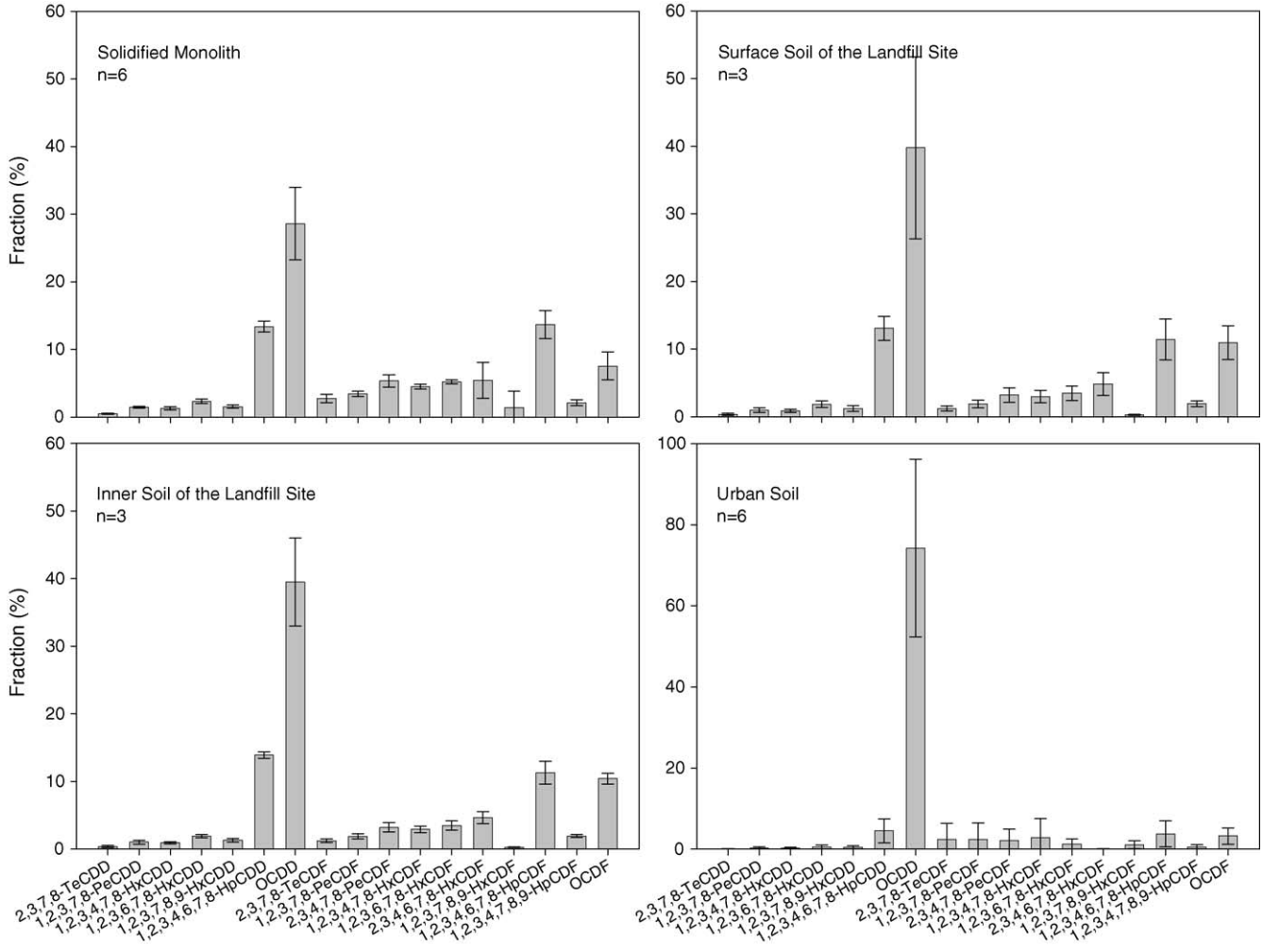

Fig. 1. The congener profiles of PCDD/Fs (mean ± S.D.) detected from the solidified monoliths and soil samples.

Table 4
PCDD/F content in the banyan leaves of urban area and the landfill site

PCDD/Fs	Urban area ($n=6$)			Landfill site ($n=6$)		
	Range	Mean	R.S.D. (%)	Range	Mean	R.S.D. (%)
PCDD (ng kg^{-1})	7.36–18.8	12.5	36.1	11.7–42.2	22.5	47.8
PCDF (ng kg^{-1})	8.96–26.1	17.0	40.9	11.9–57.6	24.1	69.48
PCDDs/PCDFs ratio	0.54–0.94	0.76	17.8	0.73–1.38	1.01	21.1
Total PCDD/Fs (ng kg^{-1})	16.3–45.0	29.5	37.6	23.6–99.8	46.6	58.2
Total I-TEQ (ng I-TEQ kg^{-1})	1.29–3.82	2.48	42.6	1.92–11.4	4.20	85.5

Table 5
PCDD/F concentrations in the water of control group, the groundwater of monitoring wells and the treated landfill leachates

PCDD/Fs	Control group			Groundwater in the monitoring well of the landfill site			Treated landfill leachates	
	Groundwater ($n=1$)	River		Upstream ($n=1$)	Midstream ($n=1$)	Downstream ($n=1$)	Mean ($n=3$)	R.S.D. (%) ($n=3$)
		Mean ($n=4$)	R.S.D. (%) ($n=4$)					
PCDD (pg L^{-1})	0.181	1.95	33.7	13.5	20.7	10.4	1.58	6.0
PCDF (pg L^{-1})	0.0889	0.364	29.0	6.43	10.9	6.47	1.36	7.0
PCDDs/PCDFs ratio	2.04	5.32	12.4	2.10	1.90	1.60	1.16	12.9
Total PCDD/Fs (pg L^{-1})	0.270	2.32	32.6	19.9	31.7	16.8	2.94	0.06
Total I-TEQ (pg TEQ L^{-1})	0.00534	0.0155	68.6	0.0493	0.159	0.0473	0.0711	97.8

respectively. The mean PCDD/F concentrations in the groundwater and river water samples, which collected from other county were 0.00534 and 0.0155 pg I-TEQ L^{-1} while that in the upstream, midstream and downstream monitoring wells of the landfill site were 0.0493, 0.159 and 0.0473 pg I-TEQ L^{-1} (Mean: 0.0852 pg I-TEQ L^{-1}), respectively. The mean PCDD/F concentrations in the groundwater of the landfill site were 16(=0.0852/0.00534) times higher than that of background sample. Furthermore, the PCDD/F concentration of the midstream monitoring well, which is beside the landfill site, is 3.2(=0.159/0.0493) times higher than that of the upstream one, but the downstream monitoring well was at the same level with the upstream one. It suggested that the solidification could not stop PCDD/Fs leaching from the fly ash, however, the pollution region is mostly confined around the landfill site because of the strong hydrophobicity and insolubility of PCDD/Fs.

The mean PCDD/F concentration in the treated landfill leachates was 0.0711 pg I-TEQ L^{-1} (Range: 0.0219–0.120 pg TEQ L^{-1}, R.S.D.: 97.8%) and was higher than that of Behnisch et al. [14], who investigated one landfill leachate treatment plant in Japan and reported that the PCDD/F concentrations of untreated and treated landfill leachates were 0.124 and 0.027 pg WHO-TEQ L^{-1}, respectively. Comparison with the river water, the PCDD/F concentration in the treated landfill leachates was about five times higher. It revealed that besides the wet and dry deposition, stormwater runoff erosion and discharges of anthropogenic wastes, the treated landfill leachates could be a PCDD/F source of nearby water environment.

4. Conclusions

Removing PCDD/Fs from flue gas by activated carbon is merely transferred PCDD/Fs to fly ash. For preventing hazard caused by the PCDD/Fs in the fly ash, Taiwan government regulated that fly ash of MSWIs must be solidified by cement and then evaluated by TCLP to forward to the landfill sites for solidified monoliths of fly ash. However, we found that although the PCDD/F leaching concentrations of the solidified monoliths of the fly ash were extremely lower than the Taiwan PCDD/F TCLP regulation of solidified monoliths, the PCDD/F contents in the surface soils of the landfill site are much higher (460-folds) than that of urban soils and the highest value obtained in this study is 2.8 times higher than the Taiwan soil regulation (1000 ng I-TEQ kg^{-1}). It reveals that the impropriety of using solidification for final treatment of fly ash and the need to reconsider the PCDD/F regulation of the solidified monoliths of the fly ash. The PCDD/F concentrations in the groundwater and the treated landfill leachates of the landfill site for solidified monoliths were both higher than that in the control samples, suggesting its potential to be a PCDD/F source of nearby water environment. Although landfill sites for solidified monoliths of fly ash are not included in the PCDD/F inventory because they do not involve original releases, however, if without proper control and management, they can seriously hazard not only the surrounding environment but also people who live far away because PCDD/Fs may undergo atmospheric transport and deposit in such distant areas and, therefore, are important to consider.

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