CHARACTERISTICS OF DIOXIN-LIKE COMPOUNDS IN LEACHATES FROMLANDFILLS CONTAINING INCINERATION RESIDUES IN TAIWAN

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Abstract

Total contents of PCDD/Fs and dioxin-like -PCBs in soluble phase and suspended particulate matter (SPM) of leachate, before and after the leachate treatment process from landfills containing incineration residues in Taiwan were determined. The total contents of PCDD/Fs and dioxin-like -PCBs ranged from 0.0585 to 3.23 pg TEQ/L and 0.00300 to 0.431 pg TEQ/L, respectively. Levels of PCDD/Fs and dioxin-like PCBs in treated leachate were much less than that in raw leachate, with the exception of the sample collected from landfill A. The results obtained from this study indicated that dioxin-like compounds can be removed through the process such as coagulation, aeration, sedimentation, filtration, biological treatment and activated carbon absorption in selected landfills. The dominated PCDD/Fs in leachate was 1,2,3,4,6,7,8-HpCDD and the removal rate for leachate liquid was 5.70～95.6% and 41.2～97.6% for leachate solid. As for the dioxin-like PCBs, the most dominated PCB congeners in leachate liquid and solid were PCB 126, 169 and 118. The removing rate of PCBs for leachate liquid ranged from of 39.5 to 99.1% for landfills C, D and E whereas 34.1～99.6% for leachate solid in landfills B,C,D,E, and F. The results obtained from this study confirmed that the concentrations of PCDD/Fs in selected landfill sites for co-treated solidified fly ash and bottom ash were not particularly higher than the other landfills, furthermore, it was lower than the Taiwan PCDD/Fs TCLP regulation of solidified monoliths. Nevertheless, the potential source of dioxins from the solidified fly ash that leaks into the surrounding soil environment need to be further addressed.

Introduction

Although the distribution of dioxin-like compounds such as polychlorinated dibenzo-p-dioxins/ dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in various environmental media produced from different emission sources had been widely discussed. The survey on PCDD/Fs leaching concentrations and characteristics of co-existing compounds on PCDD/Fs and PCBs in landfills treating incineration residues are currently very limited in Taiwan[1]. Many of the studies indicated that the municipal solid waste incinerator (MSWI) has been considered as the main MSW treatments which are accounted for the major dioxin like chemicals emission sources[2]. The fly ash generated from MSWI must be stabilized or solidified before disposed to the landfill sites due to its high contents of dioxin-like compounds, heavy metals and other possible potential carcinogens. Therefore, the leaching characteristics of these incineration residues have become an important issue. The first survey of the concentrations of PCDD/Fs and dioxin-like PCBs in leachate samples before and after leachate treatment plant from six landfill sites in Taiwan was conducted in this study. The objectives of the current investigation were to reveal the concentrations of PCDD/Fs and dioxin-like PCBs in raw and treated leachates and to examine the removing efficiency of PCDD/Fs and dioxin-like PCBs in selected landfills.

Materials and Methods

Twelve samples were collected from six landfill sites throughout Taiwan. Landfill Shulin (A), Shanzhuku (B) and Bali (C) landfills, Taichung City (D), Tainan City (E) and Kuoshiung City (F), are distributed in northern, central and southern part of Taiwan, respectively (Fig 1). The descriptions of the selected six landfills are summarized in Table 1. All the landfills are still in operation and mainly treating bottom ash or co-treated solidified fly ash, however, municipal solid wastes were dumped into the landfills in the initial stages. The samples composed of liquid and suspended solid phase were collected before and after the leachate treatment process. In order to obtain sufficient amount of analytes, the leachate samples (20-48L) were collected using the on-site large volume pre-concentration system (Fig. 2). The system was equipped with fiber filter (0.5μm pore size) /case for collecting particle-bound PCDD/Fs, polyurethane foam (PUF) /holder to retain the target compounds in the liquid phase, an air bubble removal device, vacuum pressure sensor and computer panel. The PUF and filter samples were then soxhlet extracted (24h, extracted by toluene) and silica gel clean-up procedure. The samples were fortified with internal standards (6 ^{13}C-PCDDs, 9 ^{13}C-PCDFs and 12 ^{13}C-dioxin-like PCBs) before extraction. A CAPE carbon column was used to separate interferences, PCDDs/Fs and dioxin-like PCBs. Dioxin-like PCBs portion were eluted from carbon

column in forward direction with 6 mL of hexane/toluene, and then PCDDs/PCDFs fraction was eluted from carbon column in reverse direction with 35 mL toluene. Before instrument analysis, ^{13}C-labeled standards were added and the vials were vortexed to mix completely. All analyses were performed with the isotope dilution method. Finally, the PCDD/Fs and DL-PCBs were analyzed by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS, HP 6890/JEOL JMS-700), equipped with positive electron impact (EI+) source. A DB-5 MS column (L=60m, i.d.=0.25mm, film thickness=0.25 μm, carrier gas helium, J&W Scientific) was employed with the following temperature program: 150°C for 3 mins, increased to 210°C at a rate of 30°C /min for 15 mins and then increased to 230°C at 1.5°C /min and finally to 310°C at 15°C /min. All measurements were made in selective ion recording (SIR) mode had a resolving power of 10,000 and two most intense ions of the molecular ion cluster. The details of the quality control were as described in the EPA method 1613B and 1668A. Toxicity equivalent (TEQ) concentrations were calculated by using the WHO2005 equivalency factors (WHO-TEFs).

Results and Discussion

We collected the raw and treated leachates from the landfills in order to determine the levels of PCDD/Fs and dioxin-like PCBs and examine the removal efficiency of leachate treatment process in Taiwan. The results showed that the total PCDD/Fs concentration in collected samples ranged from 0.0585 to 3.23 pg TEQ/L. The highest PCDD/Fs concentration in raw leachate was found at landfill B (3.23 pg TEQ/L) followed by landfill E (1.48 pg TEQ/L). However, the relationships of the size between landfills and PCDD/Fs concentrations were not significant. Fig 3 and 4 illustrated the concentrations of 17 PCDD/Fs congeners in raw leachate between liquid and solid phases were ranged from 0.0348 ～ 1.24 and 0.0549 ～ 3.17 pg TEQ/L with the highest concentration was 1,2,3,4,6,7,8-HpCDD. In addition, Fig 5 and 6 demonstrated the highest concentrations of PCDD/Fs in treated leachate liquid and solid phases was 2,3,4,7,8-PeCDF, respectively. It was obvious that the solid phases PCDD/Fs in leachate played a major role as expected. The removal efficiency of PCDD/Fs for leachate between liquid and solid phases were varied among 5.70～95.6% and 41.2～97.6%, respectively, except landfill A exhibited minus removal efficiency. The results also indicated that over 90% of hepta- (landfills B,C,D,E) and penta-(landfill F) substituted PCDD/Fs in solid-phase were effectively removed by the leachate treatment process in landfill leachates whereas the levels of 17 PCDD/Fs were increased in landfill A. Total PCDD/F concentrations measured in six landfill sites in Taiwan were significantly lower than those reported in Korea leachates [3,4] (11.34; 4.1~6.22 pg TEQ/L) but similar to those of Japan landfills (3.83 pg TEQ/L)[5]. As for the dioxin-like PCBs, the total concentrations were in the range of 0.00300 to 0.431 pg TEQ/L. The concentrations of 12 dioxin-like PCBs in raw leachate between liquids and solids phase were ranged from 0.00273-0.271 and 0.00452~0.342 pg TEQ/L, respectively, and PCB 126 and 77 were the dominated congener in landfills E and B, as shown in Fig. 7 and Fig. 8. Furthermore, the concentrations of 12 dioxin-like PCBs in treated leachate between liquid and solid phases were ranged from 0.00148~0.0169 pg TEQ/L and 0.000589~0.0441 pg TEQ/L, respectively. The data shown in Fig.9 and Fig. 10 described the concentration of PCB 126 was the highest in landfills A and C. The removing efficiency (efficiencies?) of PCBs for leachate liquid were in the range of 39.5 ～99.1% for landfills C,D and E while 34.1～99.6% for leachate solid for landfills B,C,D,E, and F. The three most dominated PCBs detected were PCB126, 169 and 118 in both leachate liquid and solid samples. The current investigation identifies the *non-ortho* PCBs comprising up to 90.3% in leachate liquid from landfill F and 96.7% in leachate solid sample collected from landfill D, respectively. However, among these three PCB congeners, the highest removal efficiency was found in PCB118 which represented 25.3～99.6% in the leachate liquid and 30.9～99.9% in the leachate solid. In conclusions, the results provided thoroughly information regarding the proper MSW treatment for the safety of the incineration residue disposal to the landfill.

Acknowledgements

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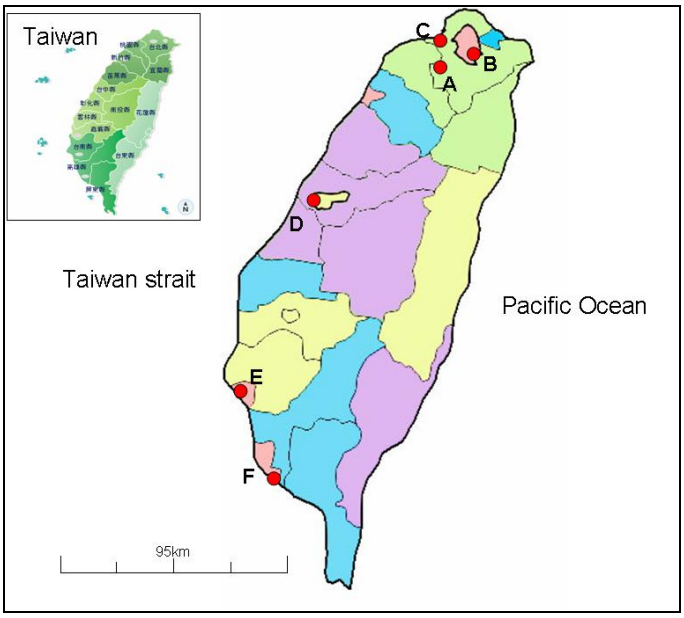

Fig 1. Sampling sites from six landfills in Taiwan (A: Shulin Landfill; B: Shanzhuku Landfill; C: Bali Landfill; D: Taichung City Landfill; E: Tainan City Landfill; F: Kaohsiung City Landfill).

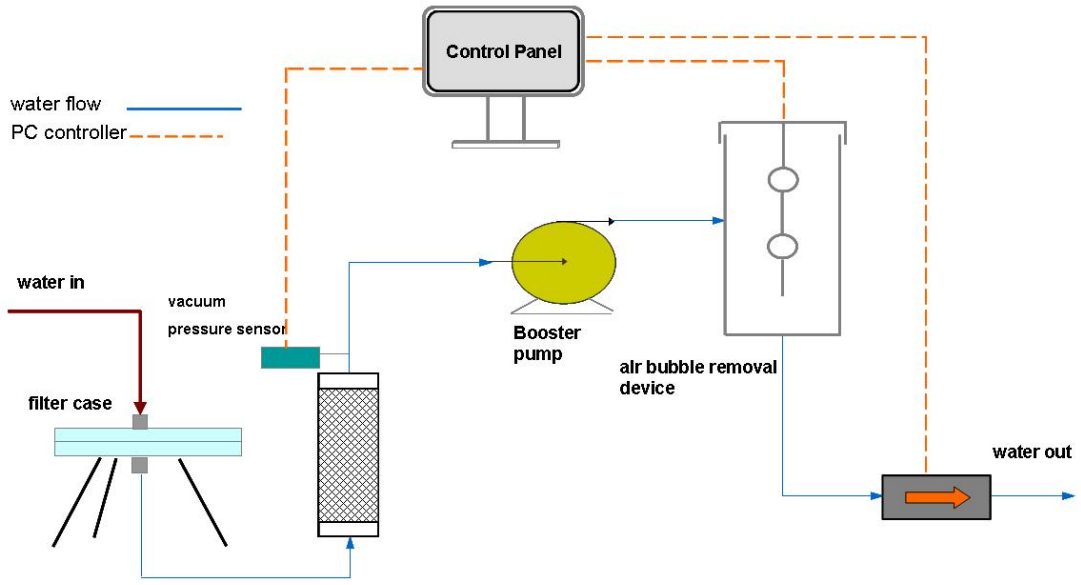

Fig 2. Diagram of the large volume on-site pre-concentration system.

Table 1. Descriptions of selected landfills in Taiwan

Landfills	A	B	C	D	E	F
Operation Year	1996/3~	1994/6~	2000/1~	1998/12~	2002/1~	1999/2~
Condition	Open	Open	Open	Open	Open	Open
Area (ha)	36.4	30	27.6	12.07	53.5	20
Available Capacity (M^3)	217373	280000	387046	23000	30000	400000
Designed Leachate Volume (CMD)	500	1000	800	450	800	130
Accepted waste type	1. bottom ash 2. solidified fly ash	bottom ash	bottom ash	bottom ash	bottom ash	1. bottom ash 2. solidified fly ash

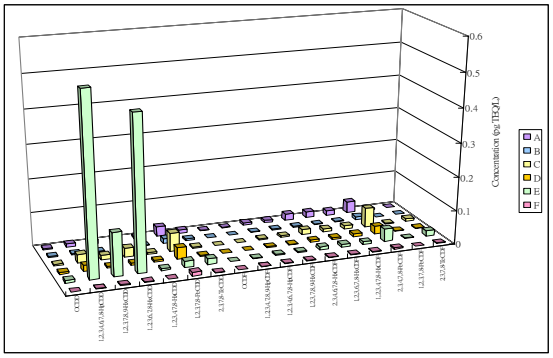

Fig 3. Congener profiles of 17 PCDD/Fs in raw leachate liquid phase.

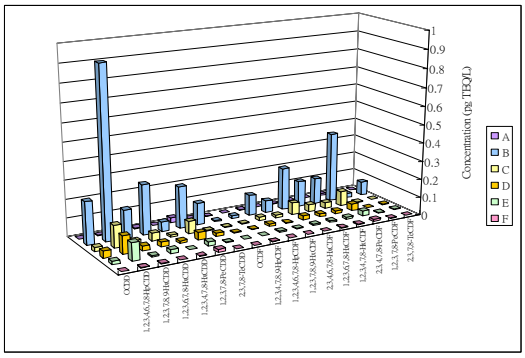

Fig 4. Congener profiles of 17 PCDD/Fs in raw leachate solid phase.

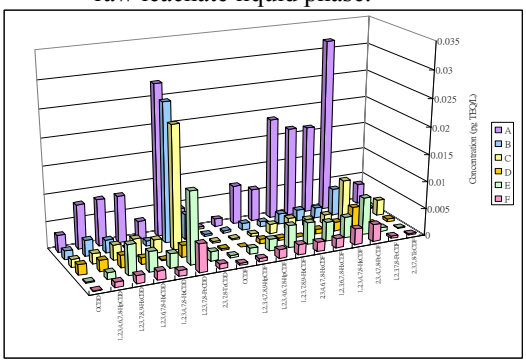

Fig 5. Congener profiles of 17 PCDD/Fs in treated leachate liquid phase.

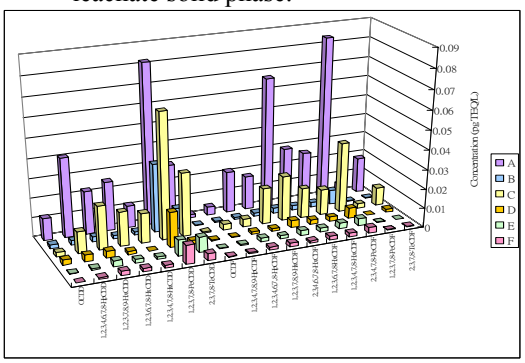

Fig 6. Congener profiles of 17 PCDD/Fs in treated leachate solid phase.

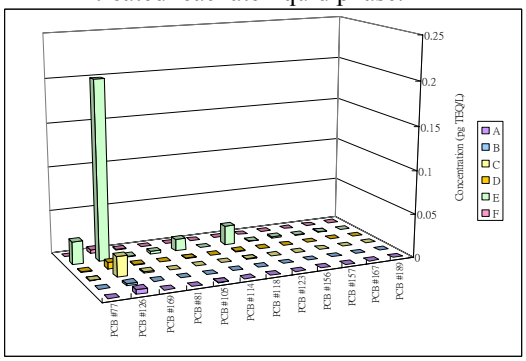

Fig 7. Congener profiles of 12 dl-PCBs in raw leachate liquid phase.

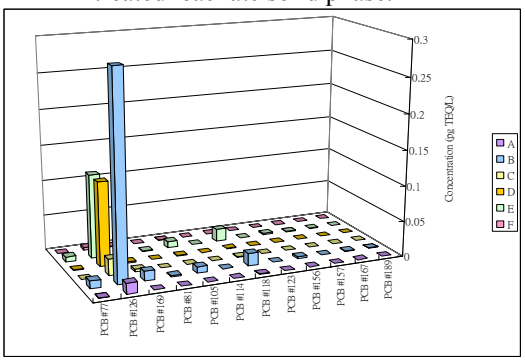

Fig 8. Congener profiles of 12 dl-PCBs in of raw leachate solid phase.

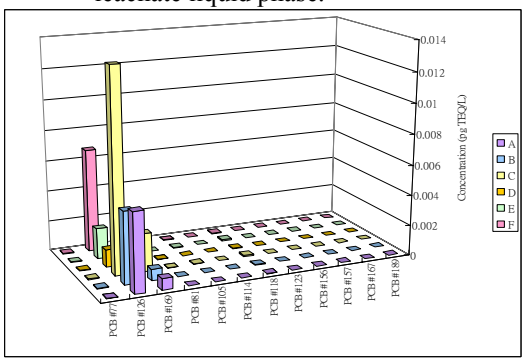

Fig 9. Congener profiles of 12 dl-PCBs in treated leachate liquid phase.

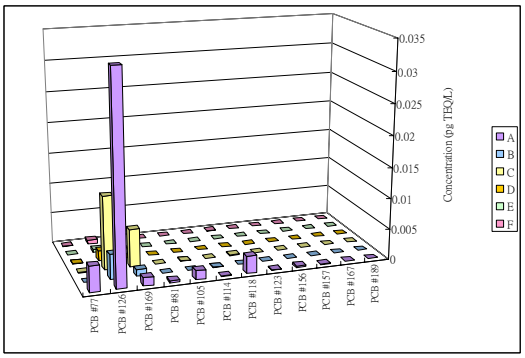

Fig 10. Congener profiles of 12 dl-PCBs in treated leachate solid phase.