

Levels of PCDD/Fs in Soils in the vicinity of the Municipal Solid Waste Incinerator in Shanghai, China

Lijuan Jia^{1*}, Yunyun Deng², Wanlian Mao³, Haowen Yin⁴, Feng Tao⁵, Fang Huang⁶

^{1,2,3,4,5,6} Bioassay and Safety Assessment Laboratory, 1500 Zhangheng Road, Zhangjiang Hi-tech Park, Shanghai 201203, P.R.China

Introduction

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/F) are two groups of semi-volatile organic contaminant that are extremely toxic and can accumulate in organic carbon-rich media such as soil and sediment, due to their hydrophobic properties. Many studies have evaluated the PCDD/F levels near emission sources and their impact on the environment¹⁻⁴. In China, the incineration of municipal, industrial and medical waste is considered to be a significant source owing to the rapid increase in the use of incineration for waste disposal in China⁵⁻⁶. The studied municipal solid waste incinerator(MSWI) is located in the Pudong area with a daily capacity of 1,000 tons. It was the first kiloton sized modern garbage incineration plant in China, and officially began operation in July 2003. To generate overall information on the environmental impact of municipal solid waste incinerator(MSWI) in Shanghai, this study investigated the levels of PCDD/Fs in agricultural soils near the MSWI in Shanghai, China.

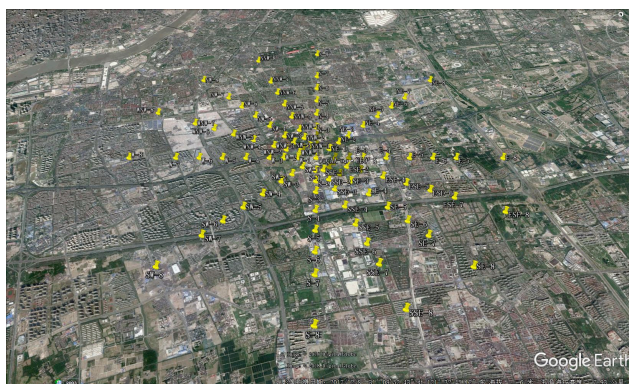


Fig.1 Distribution of sampling sites in soils around the municipal waste incinerator in Shanghai

Materials and methods

Materials

n-Hexane(pesticide) and acetone(pesticide) were obtained from Fluka, USA and dichloromethane (DCM, pesticide) were from J.T.Baker, USA. Silica gel(0.063–0.200mm) and Florisil(0.150~0.250mm)was supplied by Merck, Germany. Anhydrous sodium sulphate (GR) from Shanghai Pharmaceutical Group was baked at 450℃ before use and stored in sealed containers. Nonane (p.a.) was purchased from Fluka, USA. The standards of PCDDs were supplied by Wellington Laboratories, USA.

Sampling and sample preparation

The soil samples were collected during spring (from April to May) in 2017, with a total of 60 soil samples collected within a 3km radius of the municipal solid waste incinerator (MSWI), mainly in the prevailing year-

round downwind directions (E, ESE, SE, SSE, W, WNW, NW, NNW) (Fig. 1). In each direction, one sampling site was set every 300m in the first 1500m from the incinerator, every 500m from 1500–3000m. Every site was located using the global positioning system (GPS). The sampling locations were on open fields, not excessively covered by crops. Three samples of top soil were taken within a 5–10 m radius of each sampling site at a depth of 0–20 cm using a pre-cleaned steel spoon. The three samples were then mixed in a glass bottle and transported to the laboratory. The samples were dried at room temperature for 10–14 days. Dried soils were crushed by a ceramic cutting mill and then passed through a stainless steel sieve with a mesh size of 1×1 mm. The moisture level of these soil samples was less than 3%. The samples were then labeled and stored in glass flasks at a temperature of –20°C until analysis.

Analytical methods

PCDDs were extracted from the soil samples using ASE 200 system (Dionex, USA). 10g soil samples (dry weight) were filled into 22 ml extraction cells and spiked with a mixture of ¹³C-labeled compound stock solution (10 µl) before extraction. The samples were extracted using 1:1 mixture of hexane and dichloromethane () followed by clean-up with multilayer silica gel column and florisil column according to USEPA Method 1613b. High resolution gas chromatography-high resolution mass spectrometry(HRG-HRMS) was undertaken on an Agilent 6890 gas chromatograph coupled with an Autospect Premier mass spectrometer (Waters) running with an EI source in SIM mode. The recoveries generally varied between 40% and 130%, which satisfied the requirements of USEPA Method 1613b. PCDDs TEQ calculation was done using I-TEFs.

Results and discussion

Levels and congeners distribution

Table 1 summaries the 2,3,7,8-substituted PCDD/Fs concentrations of the 60 soil samples collected around MSWI. The total PCDD/F concentrations of the soil samples ranged from 41.9 to 2149 pg g⁻¹ (0.39–7.16 pg I-TEQ g⁻¹), with a mean of 216.4 pg g⁻¹(2.19 pg I-TEQ g⁻¹). Compared with domestic and foreign soil average PCDD/F levels, the I-TEQ values observed in the present study were much lower than those from Spain (1.22–34.28 pg I-TEQ g⁻¹)², Finland (13–252 pg I-TEQ g⁻¹), England (2–272 pg I-TEQ g⁻¹)⁷ and Korea (1.25–74.98 pg I-TEQ g⁻¹)¹, which were also near MSWIs. The PCDD/F concentrations in soil samples from Shanghai, Japan(average 7.10 pg I-TEQ g⁻¹)¹ and the United States(average 4.0 pg I-TEQ g⁻¹)⁹ are of the same order of magnitude and are comparable with the soil guidelines in Germany and Sweden, which are 5 and 10 pg I-TEQ g⁻¹, respectively. Although no great variation in the PCDD/F concentrations was observed for most of the soil samples, unusually high values for TEQ were found in four soil samples (SSE2, ESE2, W6, ESE2), with concentrations of 7.16 pg I-TEQ g⁻¹, 6.96 pg I-TEQ g⁻¹, 6.56 pg I-TEQ g⁻¹, 5.86 pg I-TEQ g⁻¹ respectively. These observed PCDD/F concentrations all exceed the 5 pg I-TEQ/g⁻¹ limit which restricts the cultivation of certain vegetables. According to the soil dioxin guideline concentrations of Germany, this soil should be limited to cultivation of plants with minimum dioxin transfer, e.g., corn and soybeans.

Table 1 PCDD/F concentrations and I-TEQ of the agricultural soils in vicinity of the incinerators (pg g⁻¹)

Distance(m)	E		ESE		SE		SSE					
	ΣPCDD/Fs	I-TEQ	ΣPCDD/Fs	I-TEQ	ΣPCDD/Fs	I-TEQ	ΣPCDD/Fs	I-TEQ				
300	E1	80.4	0.39	ESE1	304.8	3.91	SE1	483.2	3.09	SSE1	122.4	1.29
600	E2	140.3	1.76	ESE2	1273	6.96	SE2	126.5	3.14	SSE2	2147	7.16
900	E3	179.0	2.56	ESE3	147.2	5.86	SE3	149.4	1.92	SSE3	108.1	2.06
1200	E4	107.2	1.16	ESE4	191.8	2.05	SE4	175.6	1.86	SSE4	166.6	1.55

1500	E5	91.8	1.09	ESE5	126.2	1.93	SE5	155.4	1.95	SSE5	121.3	0.95
2000	E6	206.7	1.65	ESE6	864.2	1.78	SE6	68.3	0.90	SSE6	237.9	2.23
2500	E7	164.2	0.90	ESE7	201.1	3.05	SE7	75.2	1.10	SSE7	223.1	3.59
3000	E8	559.4	3.24	ESE8	192.4	2.14	SE8	114.8	1.53	SSE8	137.8	1.45
Distance(m)	W		WNW		NW		NNW					
	Σ PCDD/Fs	I-TEQ	Σ PCDD/Fs	I-TEQ	Σ PCDD/Fs	I-TEQ	Σ PCDD/Fs	I-TEQ	Σ PCDD/Fs	I-TEQ		
300	W1	132.5	1.34	WNW1	150.3	0.62	NW1	92.6	0.95	NNW1	134.7	0.62
600	W2	147.1	2.24	WNW2	175.1	2.22	NW2	131.5	1.84	NNW2	119.7	1.79
900	W3	41.9	0.50	WNW3	100.3	1.24	NW3	121.7	1.16	NNW3	95.0	1.57
1200	W4	-	-	WNW4	129.8	1.88	NW4	80.9	1.20	NNW4	141.2	2.28
1500	W5	-	-	WNW5	485.6	1.31	NW5	-	-	NNW5	120.6	1.45
2000	W6	194.8	6.56	WNW6	67.5	1.82	NW6	196.5	2.64	NNW6	99.1	1.35
2500	W7	122.6	3.85	WNW7	-	-	NW7	111.1	1.44	NNW7	138.0	4.78
3000	W8	175.6	1.92	WNW8	91.8	3.09	NW8	109.6	1.19	NNW8	140.5	2.55

-: not analyzed.

Among all soil samples, OCDD was the predominant congener, accounting for around 84.35% of the total concentration of 17 congeners, followed by the highly-chlorinated congeners including 1,2,3,4,6,7,8-HpCDD and OCDF, which account for 4.50% and 3.74% respectively. This distribution profile is similar to those found in soils around MSWI²⁻⁴. With respect to the TEQ concentrations, the main contributor to the TEQ was 2,3,4,7,8-PeCDF (accounting for 33.33% of the total TEQ), which is also consistent with previous research²⁻⁴. The PCDD/F level of the background agricultural field from nine districts around Centertown in Shanghai can be found in Li et al (2009)¹⁰ and Rong et al (2010)¹¹. The concentrations of the soil samples from background area were ranged from 114.24~687.93 pg g^{-1} (0.71~7.84 pg I-TEQ g^{-1}), with an average value of 211.89 pg g^{-1} (2.20 pg I-TEQ g^{-1}). Compared with the our previous report on the background soil concentrations in Shanghai, the average concentrations in soil near the MSWI is close to the background level.

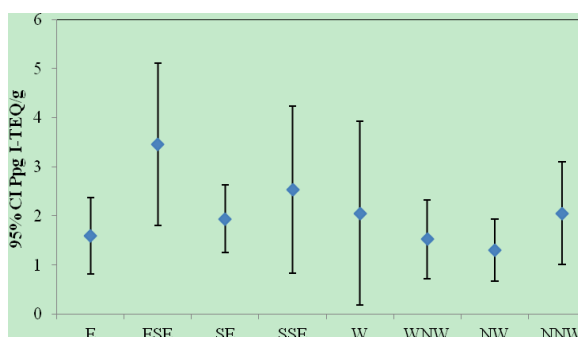


Fig.2 Error bar charts of the I-TEQ concentrations in different wind directions

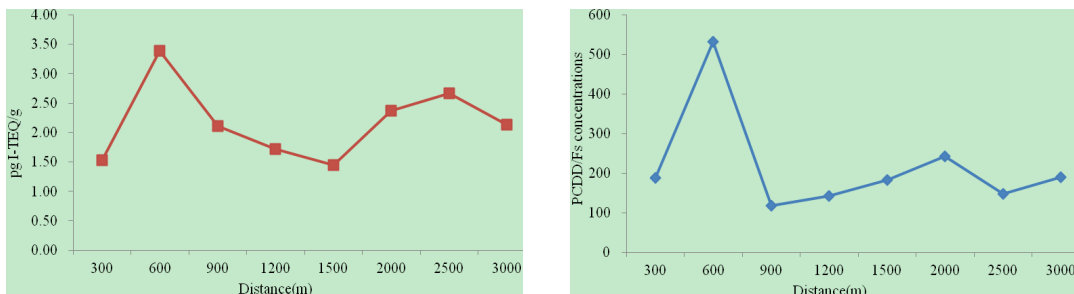


Fig.3 Trend of pg I-TEQ/g and PCDD/Fs concentration in the soils with the increasing distances

Fig. 2 represents the I-TEQ concentrations in different directions for MSWI. It shows that the TEQ values in samples from the ESE and SSE directions are higher than those from other directions. For the plot of the distance against average concentration (Fig. 3), the 60 soil samples were roughly divided into eight groups based on distance from the MSWI (300, 600, 900, 1200, 1500, 2000, 2500 and 3000m). Initially, the mean concentrations of PCDD/Fs in the soil samples increased with the distance from the incinerator until peak levels occurred, and then the concentrations decreased with further distance. The maximum PCDD/F levels were observed approximately 600m from the MSWI as the Fig. 3 showed. Incinerator fly ash will be released into the environment during the transport, processing and storage. The effect of atmospheric diffusion and dry/wet deposition will result in the presence of the peak concentrations at a certain distance (not the closest) from MSWIs, which is in accordance with the skewed or normal Gaussian model relating to the diffusion of gaseous pollutants. Generally, the concentrations of PCDD/Fs are higher in winter and the prevailing wind direction in Shanghai in winter is northwest, the soil samples collected from southeast of the incinerators (ESE, SSE) are located at the windward direction of this prevailing wind direction, which may explain why the soil samples in the ESE and SSE direction had higher concentrations than those from the other directions and showed no obvious patterns between concentration and distance.

Acknowledgements

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