

**Atmospheric bulk deposition of polychlorinated dibenzo-p-dioxins
and dibenzofurans (PCDD/Fs) in the vicinity of MSWI in Shanghai, China**

Deng YY, Jia LJ, Mao WL, Yin HW, Tao F, Huang F

Bioassay and Safety Assessment Laboratory, Shanghai Academy of Public Measurement, Shanghai, P.R.China, 201203; *Deng YY, E-mail [□dengyy@apm.sh.cn](mailto:dengyy@apm.sh.cn)

Introduction

PCDD/Fs are unwanted products of human activities, such as biomass and fossil fuel combustion, industrial processes (chemical manufacturing, metal smelting, steel sintering and paper mills, etc.), medical waste incineration (MWI), and municipal solid waste incineration (MSWI) (Kulkarni et al., 2008). For many big cities in China, in order to deal with the massive solid municipal waste, MSWIs were built in urban areas, which makes the MSWIs the important sources of PCDD/Fs in urban area. However, to the best of our knowledge, there were limited data on deposition fluxes and seasonal variations of PCDD/Fs in the vicinity of MSWI in Shanghai, China until now.

In this study, we deployed the Bergerhoff method, a glass jar to monitor PCDD/Fs at 2 sites in the vicinity of MSWI month by month for twelve consecutive sampling periods in 2017. The objectives of this study were to assess the atmospheric deposition flux and monthly variation of PCDD/Fs and to analyse possible sources of PCDD/Fs in this area.

Materials and methods

Shanghai is the political, economic, and technological center of China and is the most important region in East China. For our study area, the MSWI is located in the centre of Pudong district, which is in the southeast of Shanghai. As this plant is in close proximity to residential areas, the potential exposure of people to hazardous pollutants is a serious concern in this city. Southeasterly or northwesterly winds prevailed throughout the year in the study area, and they changed seasonally. 2 sampling sites were set in the vicinity of this MSWI. Site A was located in the northwest of the MSWI, with a distance of 900m. Site B was located in the southeast of the MSWI, with a distance of 750m.

The atmospheric deposition bulk samples were collected month by month for twelve consecutive sampling periods in 2017, using stainless steel pots with an inner diameter of 50cm and a height of 50cm, from 2 sites (Site A and Site B) in the vicinity of MSWI. The deposition samples were analyzed for 17 PCDD/Fs following US Environmental Protection Agency Method 1613b with high resolution GC-MS. The mass spectrometer had a

resolution of at least 10,000 and was operated in selected-ion-monitoring mode. The recovery ranges internal standards for PCDD/Fs were 34.3-98.1% (average \pm SD), which satisfied the requirements of the standards.

Results and discussion

The 2, 3, 7, 8-substituted PCDD/Fs deposition fluxes and TEQ fluxes are listed in Table 1. The bulk deposition fluxes of 2,3,7,8-substituted PCDD/Fs varied from 51.9 to 560 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for the Site A, and 23.5 to 440 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for the Site B. The TEQ deposition fluxes were 2.14-27.4, and 1.17-20.7 $\text{pg WHO-TEQ}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ at the Site A and Site B, respectively. There is a strong correlation between the total TEQ and 2,3,7,8-substituted PCDD/Fs with the R^2 of 0.9781. The deposition fluxes of 2, 3, 7, 8-substituted PCDD/F behaved obviously higher in June and July than other months, maybe due to wet precipitation in rainy season.

Table 1 Deposition fluxes and TEQ fluxes of PCDD/Fs in the vicinity of MSWI in Shanghai

Month	Site A		Site B	
	Deposition fluxes ($\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	Deposition fluxes ($\text{pg WHO-TEQ}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	Deposition fluxes ($\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	Deposition fluxes ($\text{pg WHO-TEQ}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
Jan	79.51	3.03	163.35	6.10
Feb	69.41	4.42	125.89	7.03
Mar	138.78	5.28	171.61	6.64
Apr	77.29	2.14	96.63	3.18
May	75.24	2.94	71.93	2.19
Jun	212.14	8.28	195.38	7.61
Jul	560.26	27.45	439.54	20.67
Aug	101.01	3.43	88.37	2.61
Sep	109.05	3.40	41.96	1.17
Oct	51.90	2.39	23.47	1.53
Nov	96.93	4.15	86.83	2.83
Dec	78.65	3.27	113.76	5.15

The dry deposition flux of total PCDD/Fs in this study was comparable to that of in the vicinity of an iron and steel making plant in Korea (204-608 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Fang et al., 2011), and in the vicinity of MSWIs in Taiwan (518-595 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Wu et al., 2009), and other areas in China, such as Guangzhou (58-900 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Ren et al., 2007). And obvious lower than the total PCDD/Fs deposition fluxes in Kanto region, Japan (1230 to 3650 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Ogura et al., 2001). Global average deposition fluxes for temperate zone was 767 $\text{pg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Brzuzy and Hits, 1996). So the dry deposition flux of total PCDD/Fs in vicinity of MSWI in Shanghai is relatively lower, comparing to other places in temperate zone.

The average profiles of 17 toxic congeners and homologues of PCDD/Fs in the bulk deposition samples are presented in Fig. 1. In general, the total homologue profiles and the 17 toxic congener profiles at two sampling sites

did not vary greatly during the twelve sampling periods. Among all deposition samples, OCDD was the predominant congener, accounting for around 40.8% of the total concentration of 17 congeners, followed by the highly-chlorinated congeners including 1234678-HpCDF, 1234678-HpCDD and OCDF, which account for 13.9%, 10.8% and 9.13% respectively. This distribution profile is similar to those found in deposition around MSWIs (Fang et al., 2011; Moon et al., 2005; Ren et al., 2007). With respect to the TEQ concentrations, 2,3,4,7,8-PeCDF is the most important contributor and accounts for 38.9% (average) of the total TEQ, followed by 234678-HxCDF and 123678-HxCDF. This result is consistent with those of previous deposition studies (Fang et al., 2011; Moon et al., 2005; Ren et al., 2007).

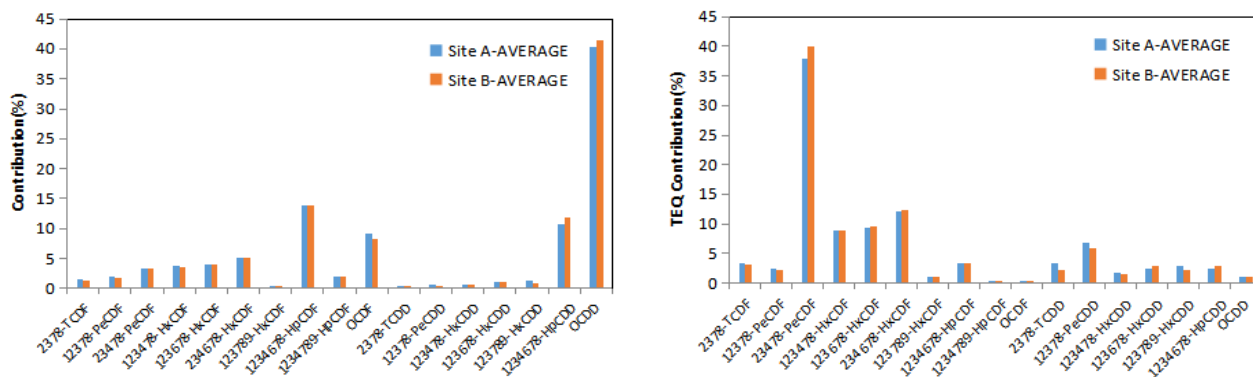


Fig. 1 Average normalized profiles of homologues and toxic 2,3,7,8-substituted congeners of PCDDs/Fs in atmospheric bulk samples from the vicinity of MSWI in Shanghai, China

For the ten homologues character, the profiles of PCDD/F homologs or congeners in the samples were the same either spatially or temporally, indicating that the PCDD/F emission sources were similar to one another. OCDD, followed by HpCDF and HxCDF, showed the highest abundance in all samples. Generally, the homologue profiles of PCDD/Fs revealed in this study were characterized by the increasing concentrations with the increasing degree of chlorination, except for OCDF. Particularly, the homologue profiles of PCDD/Fs in February and July were a little different from other months, with obviously higher HpCDF and HxCDF, but lower OCDD.

PCA has identified three main components, which account for 88.3% variability of the 24 samples, with the variables being the ten homologue families. Figure 2 shows the score plot for the PCA of PC1 and PC2. The data points for the atmosphere deposition samples near the MSWIs appear to cluster into two groups. The score plot indicates that most of the atmosphere deposition samples are in group I, which displays similar patterns to the uLGV and DV sources. The other group is clustered closer to uLGV and MSWI in the score plot, indicating this

group of samples were influenced more by the MSWIs than sampling group I. It is indicated the MSWIs were not the only emission source near the MSW incinerations and the traffic source is also an indispensable PCDD/Fs source in vicinity area of MSWI, especially for urban area.

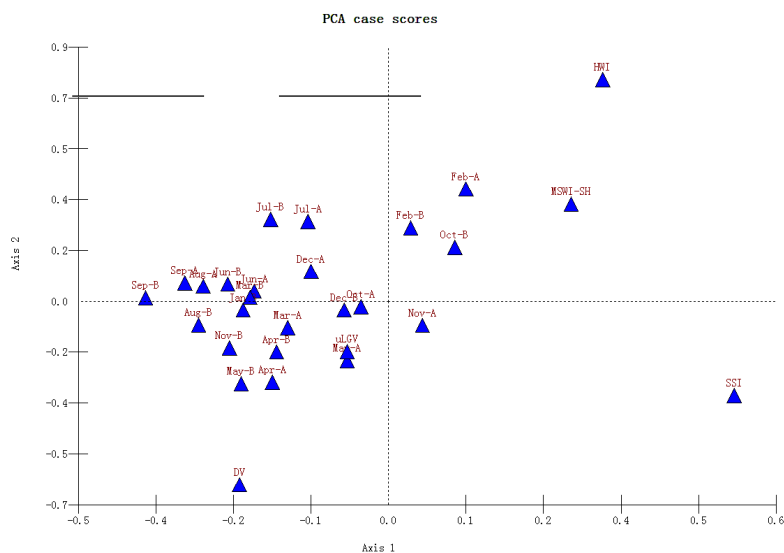


Fig. 2 Principal component plot of the 24atmosphere deposition samples and some emission sources

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