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Did municipal solid waste landfill have obvious influence on polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in ambient air: A case study in East China

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ABSTRACT

Municipal solid waste (MSW) landfill was a main way to disposal of MSW and almost 95% of MSW was disposed by landfills in the world. In order to understand the influence of MSW landfill on polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in surrounding atmosphere, 42 ambient air samples were collected and analyzed from surrounding sites, background site, upwind site and downwind site of a MSW landfill in East China. The results of present study were summarized as follows. (1) The total concentrations of PCDD/Fs (∑PCDD/Fs) in ambient air from surrounding sites, background site, upwind site and downwind site were 2.215 ± 1.004, 2.058 ± 0.458, 2.617 ± 1.092 and 1.822 ± 0.566 pg N m^{-3}, respectively. (2) The toxic equivalent concentrations (TEQ) of PCDD/Fs in ambient air from surrounding sites, background site, upwind site and downwind site were 0.103 ± 0.017, 0.096 ± 0.015, 0.120 ± 0.024 and 0.108 ± 0.014 pg I-TEQ N m^{-3}, respectively. (3) The congener profiles, ∑PCDD/Fs and TEQ between background atmosphere and surrounding atmosphere of landfill did not show statistically significant difference. (4) The ∑PCDD/Fs and TEQ in ambient air of downwind site were not higher than that of upwind site, suggesting that studied landfill did not have obvious influence on PCDD/Fs in ambient air from downwind site. (5) The 95th percentile carcinogenic risk (CR) of PCDD/Fs in ambient air from surrounding sites, background site, upwind site and downwind site were 8.03×10^{-9}, 7.57×10^{-9}, 9.69×10^{-9} and 8.15×10^{-9}, respectively, which were much lower than the threshold value of CR (10^{-6}), suggesting that studied landfill did not influence the CR of PCDD/Fs in surrounding atmosphere and negligible cancer risk occurred. (6) The non-carcinogenic risk (non-CR) analysis indicated that landfill did not have influence on the non-CR of PCDD/Fs in surrounding atmosphere and no obvious non-carcinogenic effects developed.

1. Introduction

About 0.170 billion tons of municipal solid wastes (MSWs) generated in China every year, which accounted for about 26.5% of the total MSWs in the world (Ni et al., 2009). There were a lot of MSWs treatment methods in China, including landfill, incineration and compost. As the final disposal of MSWs, landfill has been widely applied for its simple process, low investment, large handling capacity and low operating cost. In addition, some researches indicated that almost 95% of MSWs was disposed by landfills in worldwide (Ghosh et al., 2015). However, with the progress of urbanization and the residents' demands of higher living environment quality, the pollution of landfill sites for surrounding atmosphere has been attracted a growing number of attention in the world because landfill contains a great deal of potential toxic compounds and some of which may threat the safety of surrounding environment.

In recent years, some researchers studied the influence of landfill sites on groundwater quality. For instance, Talalaj (2014) analyzed the electrical conductivity (EC), total organic carbon (TOC), pH, heavy metals and polycyclic aromatic hydrocarbons (PAHs) in groundwater near a landfill site in Poland. Some researchers researched the influence of landfill sites on surrounding atmosphere. For example, Majumdar and Srivastava (2012) studied the volatile organic compound (VOCs) emissions from a landfill in India. Palmiotto et al. (2014) investigated toxicological risk

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and odor nuisance effects of five MSW landfill sites in Italy on surrounding environment, including polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs), dioxin-like polychlorobiphenyls (dl-PCBs), PAHs, vinyl chloride monomer, benzene and odor. Davoli et al. (2010) assessed the health risk of PCDD/Fs, vinyl chloride monomer (VCM) and PAHs in surrounding ambient air of landfills.

PCDD/Fs are one group of the most toxic compounds. Due to their carcinogenic and mutagenic properties, PCDD/Fs have been widely researched in various environmental media in the world, including soils, water, ambient air, food, fly ash and biota (Fernandez-Gonzalez et al., 2015; Roscales et al., 2016; González-Barreiro et al., 2015; Squadrone et al., 2016; Domingo and Nadal, 2016; Jeong et al., 2016; Perelló et al., 2015). However, study about PCDD/Fs in surrounding atmosphere of landfill was quite limited in China. In addition, existing researches about PCDD/Fs in surrounding atmosphere of landfill did not compare the congener profiles, toxic equivalent concentrations (TEQ) and total concentrations of PCDD/Fs ($\sum$PCDD/Fs) in ambient air between surrounding sites and background site and between upwind site and downwind site of landfill, and thus the influence of landfill on PCDD/Fs in surrounding atmosphere is still unknown. For example, Palmiotto et al. (2014) found that the TEQ of PCDD/Fs and dl-PCBs in surrounding atmosphere of a MSW landfill in Italy ranged from 3.0×10^{-6} to 2.0×10^{-5} pg WHO-TEQ m^{-3}; the carcinogenic risk (CR) and non-carcinogenic risk (non-CR) of PCDD/Fs and dl-PCBs in surrounding environment of the MSW landfill for residents around the landfill were, 2.2×10^{-9}–1.8×10^{-8} and 6.1×10^{-6}–9.1×10^{-5}, respectively, suggesting that the health impact of PCDD/Fs and dl-PCBs for residents were negligible. Davoli et al. (2010) performed an integrated risk assessment study in an area within 5 km from a landfill in Italy which accepts non-hazardous waste. And found that the non-CRs of PCDD/Fs were 7–9 orders of magnitude below 1, suggesting that no obvious adverse health effects occurred; the CRs of PCDD/Fs for children and adults varied from 5.5×10^{-12} to 2.4×10^{-10}, which were much lower than the threshold of cancer risk (10^{-6} or 10^{-5}), suggesting that potential incremental human cancer of PCDD/Fs emissions from studied landfill was limited.

In present study, 42 ambient air samples were collected and analyzed from surrounding sites, background site, upwind site and downwind site of a MSW landfill in East China. The differences of congener profiles, $\sum$PCDD/Fs and TEQ in ambient air between surrounding sites and background site and between upwind site and downwind site were compared, respectively, and the results were used to analyze the influence of landfill on PCDD/Fs in surrounding atmosphere. The CR and non-CR of PCDD/Fs in surrounding atmosphere and background atmosphere were quantitatively evaluated and the results were used to understand the health risk levels of PCDD/Fs in surrounding atmosphere of landfill and to analyze the influence of landfill on health risk of PCDD/Fs in surrounding atmosphere. The results quantitatively showed the influences and potential risks of PCDD/Fs posed by a MSW landfill, which can be helpful to predict the influence from MSW landfill sites and promote the management of landfill in China.

2. Material and methods

2.1. Sample collection

35 ambient air samples were collected from 5 sites in the vicinity of MSW landfill in East China (Fig. 1), including G1, G2, G3, G4 and G5. And the daily ability of studied MSW landfill was 2000 ton MSW. 7 ambient air samples were collected from background site (G6) where was far away from all possible PCDD/F sources (27 km for landfill and 10 km for mobile sources). The samples were collected in seven consecutive days in each sampling site (from January 27 to February 2, 2016). And the mixing conditions of atmosphere in sampling periods are summarized in Table 1. According to the prevailing winds of winter in landfill (northeast winds), G1 was located in upwind area and G4 was located in downwind area of landfill.

Air samples were collected using high-volume air samplers operating at 1.05 m^3 min^{-1}, which was equipped with a glass fiber filter (GFF, 220.3 cm × 25.4 cm, GB100R, 0.6 μm nominal rating) followed by a glass cartridge containing two polyurethane foam (PUF, 90 mm × 65 mm) plugs. Before sampling, the GFFs were baked at 450 °C for 12 h to remove potential organic compounds, and the PUFs were extracted by acetone and dichloromethane.

2.2. Sample analysis

17 PCDD/F congeners were analyzed according to the Chinese national standard method (HJ77.2-2008). And previous results suggested that the performance of the Chinese national standard method was comparable with USEPA method TO-9A and 8290A (USEPA, 1999 and USEPA, 2007). Briefly, the ambient air samples were transferred into Soxhlet extraction apparatus with toluene. The extracts were subjected to the following clean-up procedures: H_2SO_4 treatment, multi-layer silica gel column and alumina. The fractions which contained PCDD/Fs were collected and concentrated under N_2 purging. The injection standards were spiked, and the samples were re-dissolved in 1 mL of decane. PCDD/Fs analysis was performed on high resolution gas chromatography (HRGC) and high resolution mass spectrometry (HRMS).

2.3. Quality assurance (QA) and quality control (QC)

Procedural blanks and matrix spiked samples were processed and analyzed along with field samples and were used to enforce the quality of concentration data. Analysis of procedural blanks demonstrated that the analysis system and glassware did not have influence for contamination. For all the samples analyzed, 5 samples were randomly selected as parallel samples for repeatability checking. Only the relative standard deviation (RSD) values below 10% were accepted. The detection limits of 17 PCDD/F congeners were estimated from a signal-to-noise ratio of 3:1 in blank samples, and ranged from 0.0019 to 0.0069 pg N m^{-3} for HRGC-HRMS. The recoveries for 17 PCDD/F congeners were between 70% and 96% except O8CDF.

2.4. Health risk assessment

The health risk of PCDD/Fs in ambient air from surrounding sites, background site, upwind site and downwind site was calculated by new inhalation method. Compared with the old intake methodology, where inhalation rate (*IR*) and body weight (*BW*) were the key parameters in risk assessment process, the new inhalation method suggests that the amount of chemical that reaches the target site through inhalation, is directly related to the exposure concentration (*EC*), rather than a simple function of *IR* and *BW* (Vilavert et al., 2014). The CR of PCDD/Fs in ambient air was calculated by Eqs. (1) and (2) (Vilavert et al., 2014; Li et al., 2016a).

$$EC_{air} = \frac{C_{air} \times ET \times EF \times ED}{AT \times 365 \times 24} \quad (1)$$

$$CR_{air} = EC_{air} \times IUR \quad (2)$$

where C_{air} is the sum of converted PCDD/Fs concentrations for 17 carcinogenic PCDD/Fs in outdoor air based on toxic equivalents (I-

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