Aerosol and Air Quality Research, 18: 734–748, 2018
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ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2017.12.0587

Health Risk Assessment and Correlation Analysis on PCDD/Fs in the Fly Ash from a Municipal Solid Waste Incineration Plant

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ABSTRACT

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in fly ash may pose health threats to onsite workers due to their inevitable dispersion in the working environments during recycling and disposal of fly ash from Municipal Solid Waste Incinerators (MSWIs). Here, PCDD/Fs in fly ash from an MSWI in Southern Taiwan was analyzed from several perspectives. The results can be summarized as follows: (1) Through multiple comparison analyses, it was revealed that four types of congeners (OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF) have significantly higher concentrations than other species ($p < 0.01$). (2) 2,3,4,7,8-PeCDF represented the main contributor to the total toxic equivalent concentration (TEQ). The top three candidate indicators of total TEQ are OCDF, 1,2,3,4,6,7,8-HpCDF and 2,3,4,7,8-PeCDF, in which OCDF might be most powerful indicator of fly ash from similar sources. (3) It was indicated that all congeners correlated positively with each other (with R^2 values in the range between 0.707–0.939); Meanwhile, the results of the cluster analysis unveiled the specific features of several congeners (such as 1,2,3,7,8,9-HxCDF, OCDD and OCDF). (4) Through assessing health risk with a Monte Carlo simulation, both the 95th percentile carcinogenic risk (CR) and non-carcinogenic risk (non-CR) for onsite workers exceeded the threshold limit and should be considered as significant risks for onsite workers. (5) The results of the sensitivity analysis suggested that concentration (CC) and exposure duration (ED) were the two most sensitive parameters in both the CR and non-CR assessment. The above findings could be useful for improving existing health risk mitigation/management strategies for onsite workers in waste incineration plants.

***Keywords*:** PCDD/Fs; Health Risk; Monte Carlo simulation; Sensitivity analysis; Fly ash.

INTRODUCTION

Within the generic terms of dioxins, polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are a group of dangerous chemicals known as persistent organic pollutants (POPs). Among the PCDD/F homologues, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) is considered the most toxic form. It takes approximately 7–11 years to eliminate the half-life of 2,3,7,8-TCDD in humans (Mukerjee, 1998; Cole *et al.*, 2003). The US Environmental Protection Agency (USEPA) has listed dioxins as a serious carcinogen that promotes carcinogenesis (Cole *et al.*, 2003). Apart from causing cancer, a variety of negative or adverse health effects may be caused by dioxin exposure. Short-term exposure of humans to high levels of dioxins may be associated with increased risk of severe skin lesions such as chloracne and hyperpigmentation, altered liver function, and lipid metabolism. Long-term exposure may be linked with drastic weight loss, changes in activities of various liver enzymes, depression of the immune system, and endocrine- and nervous-system abnormalities (Vandenberg *et al.*, 2012; Wu *et al.*, 2014a). Perhaps because of the serious health effects caused by PCDD/Fs, the characteristics of PCDD/Fs in the environment have triggered numerous studies (Domingo *et al.*, 2001; Zhang *et al.*, 2013; Chuang *et al.*, 2015; Squadrone *et al.*, 2016; Tang *et al.*, 2017; Wang *et al.*, 2017; Xing *et al.*, 2017).

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Municipal solid waste incinerators (MSWIs) are a key source of PAHs, PCDD/Fs and PBDEs in the environment (Cheruiyot *et al.*, 2015; Cheruiyot *et al.*, 2016; Redfern *et al.*, 2017). In recent years, some techniques have been developed to eliminate the flue gas emissions of PCDD/Fs from MSWIs (Van Caneghem *et al.*, 2014; Cheruiyot *et al.*, 2016). Through active carbon powder adsorption and bag house filtration, PCDD/Fs in flue gas are almost eliminated and therefore enriched in the fly ash before release through the stack. Tang *et al.* (2013) reported that 65.3% of total dioxins produced by MSWI is discharged into the environment in the form of fly ash. Although fly ashes from MSWIs have been classified as hazardous waste due to the enrichment of dioxins and other chemicals, onsite workers in MSWIs are inevitably exposed to fly ash, due to its dispersion in the working environment during recycling and disposal. Due to long-term exposure to PCDD/Fs, the health risks for onsite workers in industrial environments are significantly higher than those for residents living in residential and rural areas (Shih *et al.*, 2008; Zhu *et al.*, 2017a, b). Hence, more attention should be paid to health risk evaluations, strengthening of occupational protection, and reduction of exposure to PCDD/Fs in the case of onsite workers in MSWIs. Moreover, fly ash has been classified as a hazardous waste based upon the pH, chemical components, and the particle size distribution of the ash (Tian *et al.*, 2012). These properties make fly ash a potential dermal and respiratory irritant. It is believed that there are three basic exposure pathways of PCDD/Fs: inhalation, ingestion, and direct contact (Rovira *et al.*, 2015). Therefore, it is vital to determine the key exposure pathway, which will help in providing specific personal protection equipment for onsite workers.

Generally, there are two main hypotheses regarding the mechanisms of both PCDD and PCDF formation during combustion (Lopes *et al.*, 2015). One mechanism is labeled homogeneous/high-temperature reactions, which mainly involve the rearrangement reactions of chlorinated precursors in gas. The other mechanism is called heterogeneous/low-temperature reactions, by which PCDD/Fs can also be formed from elemental carbon (*de novo* synthesis) (Zhou *et al.*, 2015). It is believed that PCDFs are mainly a result of *de novo* reactions, while synthesis from precursors play a role in PCDD formation (Xhrouet *et al.*, 2001). In addition, in the precursor model of PCDD/F formation, the PCDFs to PCDDs ratio has been found to be strongly dependent on the ratio of chlorinated benzenes to chlorinated phenols (Nganai *et al.*, 2014). Despite the numerous studies on PCDD/Fs, a unified consensus regarding the formation mechanisms of PCDD/Fs has not yet been reached. Therefore, systematic analyses on correlations among PCDD/F congeners and the PCDDs/PCDFs ratio may facilitate uncovering the formation of PCDD/Fs within different processes.

Thus, the present study has been carried out from three main aspects. First, to explore effective total PCDD/Fs-TEQ indicator for the real-time monitoring of PCDD/F emission, the mass concentration, TEQ, and characteristic index were analyzed. Then, correlation analyses and clustering were performed to disclose the potential relations among PCDD/F congeners. Subsequently, the carcinogenic risk and non-carcinogenic risk of onsite workers were quantitatively evaluated through a Monte Carlo simulation. Accordingly, the key exposure pathways and sensitive parameters contributing to CR and non-CR were detected. This study may thus provide useful background information for improving health risk management of PCDD/Fs in municipal solid waste incineration plants.

METHODS

Sampling

The fly ash samples were collected from an MSWI in southern Taiwan. Sampling was conducted from the same sampling point during 4 seasons, covering each month from 2009 to 2016. The average concentration of all months in a season was used as the quarterly concentration of PCDD/Fs.

Calculation of the PCDD/F Characteristic Index (DCI)

To facilitate screening for a rapid and cost-effective indicator of the toxic levels of PCDD/Fs in fly ash samples, the correlations between mass concentrations of individual congener and total PCDD/Fs-TEQ were examined. The calculational approaches to DCI and the linear equations describing correlations between congeners and total PCDD/Fs-TEQ equivalents are as follows (Chang *et al.*, 2011):

$$\mathrm{DCI} = \frac{\mathrm{C_{congener}}}{\mathrm{TEQ_{total}}} \tag{1}$$

$$\mathrm{TEQ_{total}} = a + b \times \mathrm{C_{congener}}, \tag{2}$$

where $\mathrm{TEQ_{total}}$ denotes the total I-TEQ (ng I-TEQ g^{-1}) of PCDD/Fs in each fly ash sample; $\mathrm{C_{congener}}$ represents the mass concentration (ng g^{-1}) of individual congeners; the slope of the fitted line is b, and a is the intercept (the value of $\mathrm{TEQ_{total}}$ when $\mathrm{C_{congener}} = 0$).

Health Risk Assessment

To calculate the incremental lifetime health risks of PCDD/Fs, it is necessary to estimate the chronic daily intake (CDI). Typically, PCDD/F exposure occurs through three routes, including inhalation, dermal contact, and accidental ingestion (which means unintentional ingestion). Here, the CDIs obtained through inhalation ($\mathrm{CDI_{inhalation}}$), dermal contact ($\mathrm{CDI_{dermal}}$), and accidental ingestion ($\mathrm{CDI_{ingestion}}$) are calculated as follows (Li *et al.*, 2016; Wu *et al.*, 2016):

$$\mathrm{CDI_{inhalation}} = \frac{\mathrm{C} \times \mathrm{InhR} \times \mathrm{EF} \times \mathrm{ET} \times \mathrm{ED}}{\mathrm{PEF} \times \mathrm{BW} \times \mathrm{AT}}, \tag{3}$$

where $\mathrm{CDI_{inhalation}}$ stands for the chronic daily intake associated with inhalation (mg kg^{-1} day^{-1}); C is the sum of the TEQ concentrations for 17 PCDD/Fs in individual samples of fly ash; InhR is the inhalation rate for the receptor (m^3 day^{-1});

the exposure frequency (day year^{-1}), the exposure time (h day^{-1}) and the exposure duration (year) are defined as EF, ET, and ED respectively; PEF refers to the particle emission factor (m^3 kg^{-1}); BW represents the bodyweight (kg), and AT denotes the average lifetime of onsite workers (70 years).

$$\mathrm{CDI_{dermal}} = \frac{\mathrm{C \times EF \times ED \times SA \times AF \times ABS}}{\mathrm{BW \times AT}} \times 10^{-6}, \quad (4)$$

where the chronic daily intake through the pathway of dermal contact is defined as $\mathrm{CDI_{dermal}}$ (mg kg^{-1} day^{-1}); SA refers to the skin surface area available for contact (cm^2); AF is the dermal adherence factor (mg cm^2-skin), and ABS refers to the dermal absorption factor.

$$\mathrm{CDI_{ingestion}} = \frac{\mathrm{C \times IngR \times EF \times ED}}{\mathrm{BW \times AT}} \times 10^{-6}, \quad (5)$$

where the chronic daily intake associated with accidental ingestion is defined as $\mathrm{CDI_{ingestion}}$ (mgkg^{-1}day^{-1}), and IngR is the accidental ingestion rate of fly ash for the receptor (mg day^{-1}).

Furthermore, carcinogenic risk was estimated through multiplying the exposure level by the cancer slope factor (CSF). The equation is as follows:

$$CR_i = CD_i \times CSF, \quad (6)$$

where CSF means the carcinogenic slope factor of 2,3,7,8-TCDD, and *i* represents different exposure pathways. The total carcinogenic risk for onsite workers was the sum of the risks through the three exposure routes, respectively, inhalation, dermal contact, and accidental ingestion.

In addition, as shown in Eq. (6), the potential non-carcinogenic risk through a single exposure pathway can be evaluated using the Hazard Quotient (HQ). Then, the Hazard index (HI) can be used to estimate the cumulative non-carcinogenic risk through three exposure routes.

$$\mathrm{HQ_i} = \frac{CD_i}{\mathrm{RfD}} \quad (7)$$

$$\mathrm{HI} = \frac{\sum CD_i}{\mathrm{RfD}} = \frac{\mathrm{CDI_{inhalation}} + \mathrm{CDI_{ingestion}} + \mathrm{CDI_{dermal}} / \mathrm{ABS_{GI}}}{\mathrm{RfD}}, \quad (8)$$

where RfD is the chronic oral ingestion reference dose for 2,3,7,8-TCDD (mg kg^{-1} day^{-1}), and $\mathrm{ABS_{GI}}$ is the gastrointestinal absorption factor.

Monte Carlo Simulation

A Monte Carlo Simulation is a very useful approach for quantitative health risk analyses (Cullen, 2011), by which uncertainty and variability are characterized through a process of generating random numbers to cover the entire range of exposure parameters in different scenarios. In this study, the CR and non-CR of onsite workers (adults) were evaluated and analyzed using the Crystal Ball 7.2 software, in which a Monte Carlo simulation was implemented. The number of iterations in the model was set as 100,000 based on the output stability (Driels and Shin, 2004). Moreover, a sensitivity analysis was also performed using Crystal Ball. The reports for the Monte Carlo simulation and the sensitivity analysis are provided in Table S1.

RESULTS AND DISCUSSION

Concentration Distribution of PCDD/Fs in Fly Ash

The concentration distribution of 17 PCDD/Fs congeners in fly ash are depicted in Fig. 1(a). Among the individual PCDD/F congeners, octachlorodibenzodioxin (OCDD, represented by the letter Q in Fig. 1) was found to be the dominant species during all sampling periods. Its concentration ranged from 4.53×10^{-3} to 6.75 ng g^{-1} (with a median of 4.36×10^{-1} ng g^{-1}). In contrast, the concentration of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TeCDD) was the lowest (range: 2.45×10^{-5} to 7.08×10^{-2} ng g^{-1}; median: 2.26×10^{-3} ng g^{-1}). To further test whether there were significant differences among the concentrations of PCDD/F congeners, a multiple comparison analysis (MCA) was conducted. As shown in Fig. 1(b), the results of the MCA indicated that the concentrations of PCDD/F congeners sharing the same letter in blue are not significantly different ($p > 0.05$). For instance, the PCDD/F congeners (including 2,3,7,8-TeCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,4,7,8,9-HpCDF, 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD and 1,2,3,7,8,9-HxCDD) are all marked with the letter "a" in blue, which implies that there are no statistically significant differences among these congener concentrations. Analogously, there are also no significant differences within congeners groups marked with a "b" (1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF and OCDF) and "c" (OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF), respectively. Meanwhile, the results demonstrated that the congeners marked with a "d" (OCDD) and "c" (OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF) exhibited significantly higher concentrations than the species marked with an "a" ($p < 0.01$). The sum of these four species accounted for 79.6% of the total PCDD/Fs tested.

Through investigating the emission behavior of PCDD/Fs during incineration of laboratory wastes, Liao *et al.* reported that the PCDD/F profiles from first quenching tower ash (FQA), secondary quenching tower ash (SQA), and baghouse ash (BHA) were similar, and all three types of PCDD/F mass were mainly distributed in OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF (Wang *et al.*, 2014). Obviously, these findings were in good agreement with previous studies (Zhang *et al.*, 2012; Wang *et al.*, 2014), and our results also demonstrated that the 7-Cl or 8-Cl PCDD/Fs are the main species in the PCDD/F congeners from fly ash. This probably results from the fact that, compared to 4–6 Cl PCDD/Fs, 7-Cl or 8-Cl PCDD/Fs have relatively higher boiling points and

therefore can be captured more easily by the scrubber and baghouse filter. Interestingly, in some other studies (Li *et al.*, 2016; Wu *et al.*, 2016; Wang *et al.*, 2017), other congeners such as 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDD and 1,2,3,4,7,8-HxCDF were found to be dominant. However, the contribution of these congeners in this case was trivial. This phenomenon is probably due to the variation in the feeds available to different incinerators, in which PCDD/F isomer patterns are produced by different formation mechanisms.

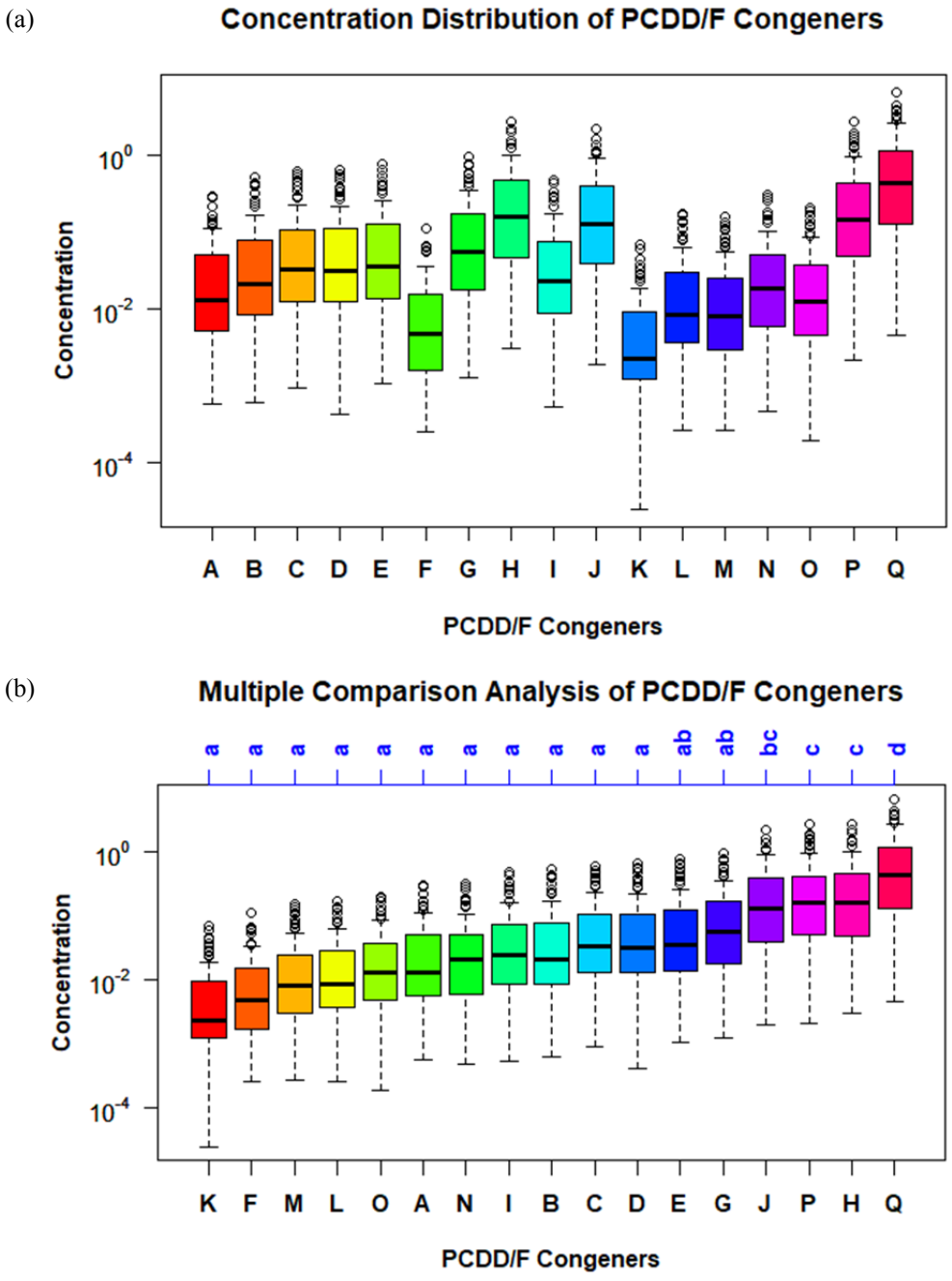


Fig. 1. (a) Concentration distribution of PCDD/F congeners. (b) Multiple comparison analysis of PCDD/F congeners. The y-axis represents the mass concentration (ng g^{-1}) of congeners; the x-axis was labeled with capital letters A–Q, which denote 17 PCDD/F congeners, respectively: 2,3,7,8-TeCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, OCDF, 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD and OCDD. The same mark rule was applied in the following text. In the boxplot, the data distribution was displayed in a standardized way based on the five-number summary: minimum, first quartile, median, third quartile, and maximum. On the top of (b), the blue lowercase letters demonstrate the results of the multiple comparison procedure. The concentrations of congeners sharing the same letters are not significantly different from each other.

The Toxic Equivalent Concentration Profiles and Characteristic Indexes of PCDD/Fs in Fly Ash

Based on the toxic equivalency factor (TEF), which indicates the degree of toxicity compared to 2,3,7,8-TCDD (Van den Berg *et al.*, 2006), the toxic equivalent concentrations (TEQ) of PCDD/Fs in fly ash were calculated. As shown in Fig. 2(a), the results suggested that total PCDD/Fs-TEQ of PCDD/Fs in a single sample ranged from 1.97×10^{-3} to 8.93×10^{-1} (ng I-TEQ g^{-1}), with a median concentration of 4.52×10^{-2} (ng I-TEQ g^{-1}). For the individual congeners, 2,3,4,7,8-PeCDF was the dominant and accounted for, on average, 35.4% of the total TEQ. However, the congener with minimal PCDD/Fs-TEQ was OCDF, which only contributed approximately 0.29% to the total TEQ. Through performing multiple comparisons among congeners, as depicted in Fig. 2(b), it was revealed that the TEQ value of 2,3,4,7,8-PeCDF was significantly higher than all other congeners ($p < 0.05$). No significant difference was detected among the TEQ values of thirteen congeners, respectively, 2,3,7,8-TeCDF, 1,2,3,7,8-PeCDF,

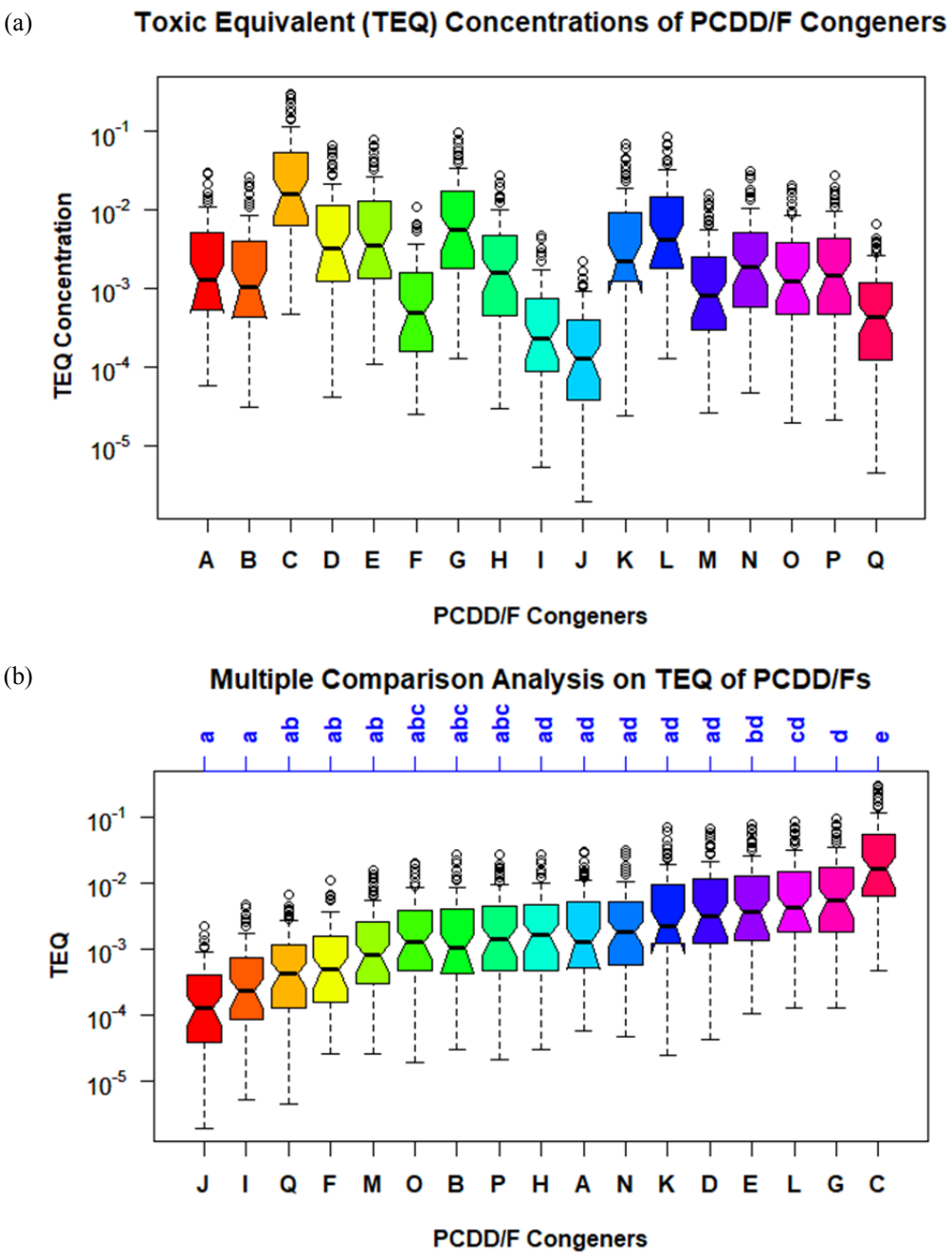


Fig. 2. (a) Toxic equivalent (TEQ) concentrations of PCDD/F congeners; (b) Multiple comparison analysis of the TEQ of PCDD/F congeners. The capital letters A-Q on x-axis denote 17 PCDD/F congeners. The y-axis represents the TEQ value of the congeners; on the top of (b), the blue lowercase letters demonstrate the results of the multiple comparison procedure. The TEQ congener values sharing the same letters are not significantly different from each other.

1,2,3,4,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, OCDF, 2,3,7,8-TeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD and OCDD. All of these 13 congeners are marked with blue letters containing "a" in Fig. 2(b). Although the concentrations of OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF were much higher than other PCDD/Fs and totally contributed about 73.4%, their total contribution to PCDD/Fs-TEQ was quite low (7.09%) due to their notably smaller toxicity factors. This result was consistent with a previous finding indicating that 2,3,4,7,8-PeCDF is the main contributor to total PCDD/Fs-TEQ (Li *et al.*, 2016; Sun *et al.*, 2017).

Recently, there were several studies on PCDD/Fs in fly ash from MSWIs. For example, Sun *et al.* (2017) reported that PCDD/Fs-TEQ varied from 7.07×10^{-2} to 7.42×10^{-1} (ng I-TEQ g^{-1}) through analyzing samples from four provinces, respectively, Henan, Anhui, Shandong, and Jilin Provinces in China. Wu *et al.* (2014b) indicated that the range of TEQ was 0.02 to 1.86 ng I-TEQ g^{-1} by characterizing the PCDD/Fs during the incineration of laboratory waste in Taiwan. Noticeably, our result was on roughly the same scale with these findings. However, Li *et al.* (2016) revealed that the TEQ was 3.2–800.1 ng I-TEQ g^{-1} by investigating the fly ash from an MSWI located in Northeast China. Liu *et al.* (2015) indicated that the PCDD/Fs-TEQ ranged from 0.005 to 87 ng I-TEQ g^{-1} through comparing PCDD/F data from 113 fly ash samples. The PCDD/Fs-TEQ levels in these results were higher than those found in our present study. It is worth noting that increasing chlorine substitution PCDD/Fs generally results in a marked decrease in toxicity. Imaginably, the observed difference in PCDD/Fs-TEQ profiles in fly ash samples are determined by distinct sources and/or incineration processes.

Furthermore, to detect which congeners can be used as effective PCDD/Fs-TEQ indicators, the DCI and Pearson correlation coefficient were calculated to measure the strength of the linear association between mass concentrations of PCDD/F congeners and total PCDD/Fs-TEQ. As listed in Table 1, the results suggested that strong positive correlations ($R^2 > 0.8$) existed between the 16 PCDD/F congeners (except 1,2,3,7,8,9-HxCDF) and PCDD/Fs-TEQ. The top two congeners with correlation coefficients (R^2) of 0.993 and 0.981 were OCDF and 1,2,3,4,6,7,8-HpCDF respectively. Perhaps because the 2,3,4,7,8-PeCDF is the major contributor to the PCDD/Fs-TEQ, it has been utilized as an indicator of the total PCDD/Fs-TEQ of the fly ash samples in previous studies (Chang *et al.*, 2011; Sun *et al.*, 2017). Here, to evaluate the efficiency of these three candidate indicators (OCDF, 1,2,3,4,6,7,8-HpCDF, and 2,3,4,7,8-PeCDF), corresponding regression equations and DCI values are illustrated in Table 1. The results disclosed that the most powerful indicator was OCDF, rather than 2,3,4,7,8-PeCDF. As is known, the toxic equivalency factor of 2,3,4,7,8-PeCDF is 0.5, which is 500 times and 50 times greater than that of OCDF (I-TEF = 0.001) and 1,2,3,4,6,7,8-HpCDF (I-TEF = 0.01), respectively. Thus, minuscule alteration/turbulence in 2,3,4,7,8-PeCDF can cause large changes in PCDD/Fs-TEQ. This can partially elucidate why the accuracy of the prediction is relatively lower when using 2,3,4,7,8-PeCDF as indicator of total PCDD/Fs-TEQ. In contrast, OCDF has not only a small I-TEF, but also significantly higher concentrations and chemical stability than other candidates. These features may improve predictive accuracy and stability. Achieving higher accuracy is crucial for prediction, particularly for real application purposes. Hence, OCDF, as a representative congener, could characterize the total PCDD/Fs-TEQ in fly ash samples from similar sources. It may be exploited as an effective total PCDD/Fs-TEQ indicator for the real-time monitoring of PCDD/F emission during the incineration process.

Correlation Analysis among PCDD/F Congeners

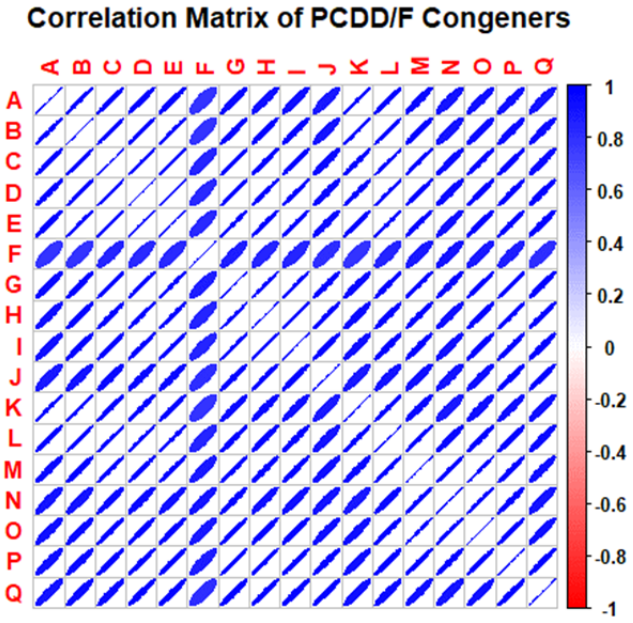
The mechanisms of PCDD/F formation, including a high

Table 1. Linear correlation between total TEQ and the mass concentrations of PCDD/F congeners (the top two are highlighted).

PCDD/F congeners	Regression equation	R^2	DCI
2,3,7,8-TeCDF	y = 0.0672 + 7.77x	0.849	0.115
1,2,3,7,8-PeCDF	y = 0.0733 + 4.64x	0.897	0.168
2,3,4,7,8-PeCDF	y = 0.0414 + 3.85x	0.932	0.243
1,2,3,4,7,8-HxCDF	y = 0.0421 + 3.75x	0.947	0.232
1,2,3,6,7,8-HxCDF	y = 0.0472 + 3.19x	0.947	0.267
1,2,3,7,8,9-HxCDF	y = 0.0875 + 23.79x	0.681	0.042
2,3,4,6,7,8-HxCDF	y = 0.0213 + 2.72x	0.967	0.353
1,2,3,4,6,7,8-HpCDF	y = 0.0183 + 0.98x	**0.981**	0.948
1,2,3,4,7,8,9-HpCDF	y = 0.0368 + 5.43x	0.967	0.162
OCDF	y = –0.0054 + 1.29x	**0.993**	0.782
2,3,7,8-TeCDD	y = 0.093 + 36.49x	0.863	0.022
1,2,3,7,8-PeCDD	y = 0.0621 + 13.18x	0.897	0.064
1,2,3,4,7,8-HxCDD	y = 0.0528 + 15.34x	0.910	0.058
1,2,3,6,7,8-HxCDD	y = 0.0482 + 7.80x	0.868	0.119
1,2,3,7,8,9-HxCDD	y = 0.0336 + 10.93x	0.915	0.085
1,2,3,4,6,7,8-HpCDD	y = 0.0237 + 1.01x	0.952	0.936
OCDD	y = –0.0002 + 0.41x	0.953	2.582

temperature homogeneous reaction and a low temperature heterogeneous reaction, are quite complicated and remain debated (Zhou *et al.*, 2015). The correlations among PCDD/F congeners and PCDDs/PCDFs ratios may be associated with the formation of PCDD/Fs within different processes. First, correlation analyses were carried out to test if there were any correlations among the concentrations of congeners (see Fig. 3). The results demonstrated that all congeners correlated positively with each other. All the average R^2 values across congeners were in the range of 0.707–0.939, with minimum and maximum R^2 values on 1,2,3,7,8,9-HxCDF and 2,3,4,6,7,8-HxCDF, respectively. Strong positive correlations among the congeners suggested their common sources and similar precursors (chlorinated aromatic hydrocarbons) (Fan *et al.*, 2017). The scatterplot of 1,2,3,7,8,9-HxCDF showed significant scatter and poor linearity, which indicated that there were differences in reaction conditions and formation processes between 1,2,3,7,8,9-HxCDF and other PCDF isomers. Moreover, it is generally agreed that there are two major pathways for the formation of polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs), namely, formation from chemically similar precursors and the so-called *de novo* synthesis (Zhou *et al.*, 2015). Imagawa and Lee (2001) proposed that 1,2,3,7,8,9-HxCDF is an indicator of the *de novo* synthesis pathway during incineration. Thus, the aforementioned phenomena, the relatively low mass concentration and low correlation coefficient of 1,2,3,7,8,9-HxCDF, may imply

(a)



(b)

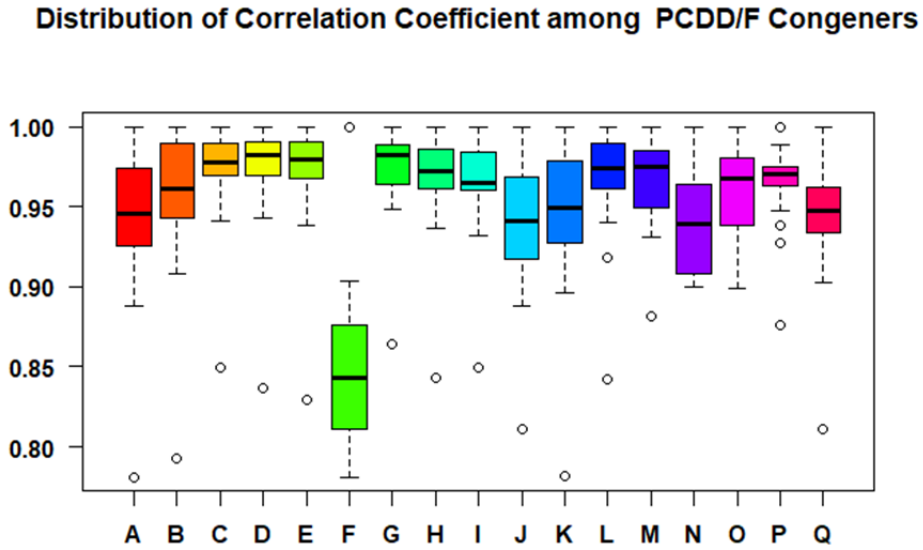


Fig. 3. (a) Correlation matrix of PCDD/F congeners. The congeners are listed down the first column and across the first row. The scatterplot in the table for the row and column intersection for two congeners demonstrates the congener pair correlations. More scatter means worse linearity. (b) The capital letters A-Q on the x-axis denote 17 PCDD/F congeners. The y-axis shows the distribution of the correlation coefficients among congeners.

that the pathway of PCDD/F formation resulting from chemically similar precursors rather than *de novo* synthesis could be the dominant one in this case.

Next, the ratio of PCDFs to PCDDs was detected based on mass concentrations. The results showed that the PCDFs/PCDDs ratios varied from 0.432 to 1.567 (with mean of 0.891). Many studies have revealed that the ratio of PCDFs to PCDDs is larger than 5 in simulated *de novo* reactions (Addink and Olie, 1995b; Chang and Huang, 2000; Pekarek*et al.*, 2001; Xhrouet *et al.*, 2001; Everaert and Baeyens, 2002). It is also believed that PCDFs are mainly a result of *de novo* reactions. Therefore, the relatively low PCDFs/PCDDs ratios suggested that *de novo* reactions may be a nonessential pathway for PCDD/F formation in this case, which further sustains the proposition that formation from chemically similar precursors is probably the major pathway in this case. Besides, in the precursor model of PCDD/F formation, the PCDFs to PCDDs ratio was found to be strongly dependent on the ratio of chlorinated benzenes to chlorinated phenols (Nganai *et al.*, 2014). PCDFs are formed predominantly from chlorinated benzenes, while chlorinated phenols are responsible for the majority of PCDDs. Thus, the observed low PCDFs/PCDDs ratio may have been mainly derived from specific features of sources.

Clustering PCDD/F Congeners across Sampling Periods

To further untangle the intrinsic relations among PCDD/F congeners, a cluster analysis of congener patterns across sampling periods was performed. A heatmap with a dendrogram, column side annotations, and row side annotations was generated (See Fig. 4). The leaves in the dendrogram on top correspond to the individual congeners. By visual inspection, PCDD/F congeners were mainly divided into five clusters, respectively marked as C1-C5 from left to right. Cluster C1 included the PCDD/F congeners: 2,3,7,8-TeCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF,1,2,3,7,8-PeCDD and 2,3,7,8-TeCDD. Cluster C2 was composed of 2,3,4,6,7,8-HxCDF, 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,7,8,9-HpCDF, which suggested that, compared to other HCDFs, these three species are more similar. As demonstrated in the top dendrogram, clusters C1 and C2 can be agglomerated into a father cluster. These two clusters suggested that 1,2,3,7,8-PeCDD and 2,3,7,8-TeCDD are two kinds of PCDDs most similar to PCDFs. Cluster C3 consisted of 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD and 1,2,3,4,6,7,8-HpCDD, which implied internal consistency among these four PCDDs. Moreover, all members of cluster C4 were OCDD and OCDF. Although, PCDDs and PCDFs are formed by divergent reaction mechanisms, cluster C4 elucidated that there are potential mutual conversion mechanisms between these two types of dioxins. Finally, the only member of cluster C5 was1,2,3,7,8,9-HxCDF. In line with our finding in the correlation analysis section, this result also implied the unique features of 1,2,3,7,8,9-HxCDF. In addition, in the dendrogram on left, the results of cluster analysis on

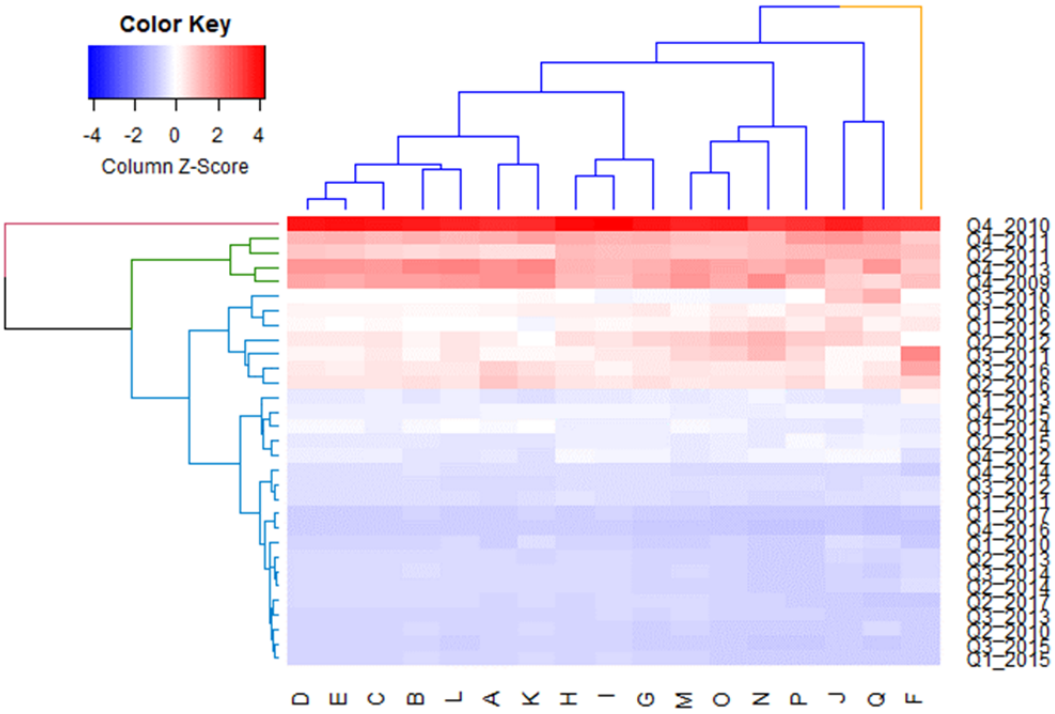


Fig. 4. Heatmap diagram of PCDD/F congener patterns across sampling periods. On the vertical margins, the dendrogram (top) and codes (below) of different PCDD/F congeners are demonstrated. The capital letters A-Q on the x-axis denote 17 PCDD/F congeners. On the horizontal margins, the dendrogram (left) and codes (right) of different sampling seasons are illustrated. Colors are coded through computing z-scores (Column scaling). The rows (columns) of the tiling are ordered such that similar rows (columns) are near each other.

samples from the four seasons (Q1, Q2, Q3 and Q4 denoting spring, summer, autumn, and winter, respectively) are illustrated. As shown, most samples from winter (such as Q4_2009, Q4_2010, Q4_2011 and Q4_2013) were grouped into one cluster. This finding may be explained partially by the fact that there is obviously a positive correlation between the removal efficiency of PCDD/Fs and temperature during the stabilization process, and the maximum removal efficiency appeared in summer, followed by spring, autumn and winter, in that order (Sun *et al.*, 2017). Thus, the results of the cluster analysis in this study can provide the finger characteristics of PCDD/Fs in the fly ash and are a useful index for different seasons.

Health Risk Assessment: Carcinogenic and Non-Carcinogenic Risk Assessment

Carcinogenic Risk Assessment

It is commonly thought that onsite workers are exposed to PCDD/Fs mainly through three routes of exposure: accidental ingestion, inhalation, and dermal contact (Li *et al.*, 2016). The total CR values of PCDD/F exposure through these three pathways were estimated and are shown in Fig. 5(a). The total CR of PCDD/Fs ranged from 5.65×10^{-9} to 36.7, with a mean value of 9.85×10^{-3}. Given the fact that simulations try to cover all possible scenarios, some of the highest values of parameters may be overestimated during the risk assessment processes. The 95th percentile

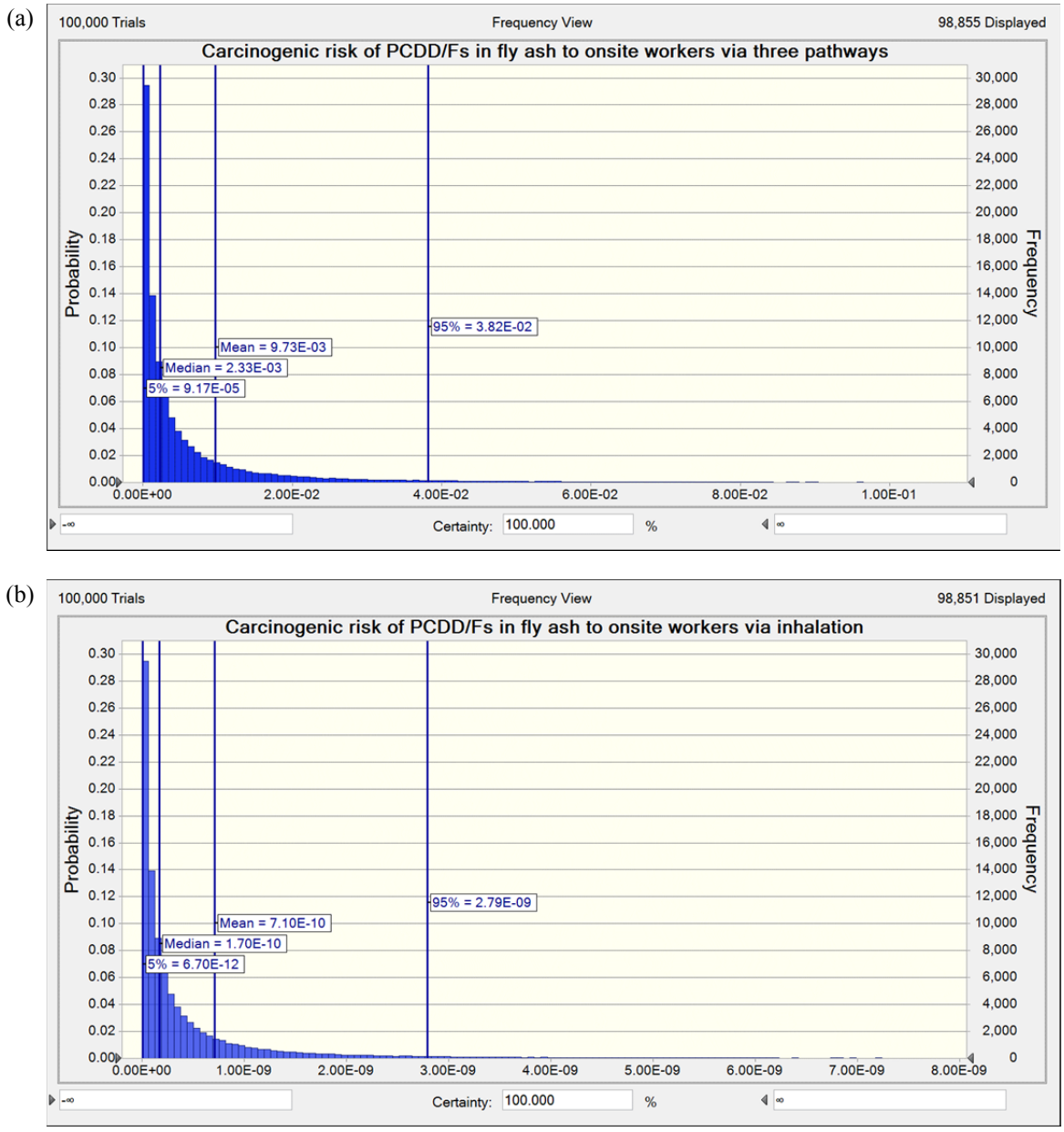
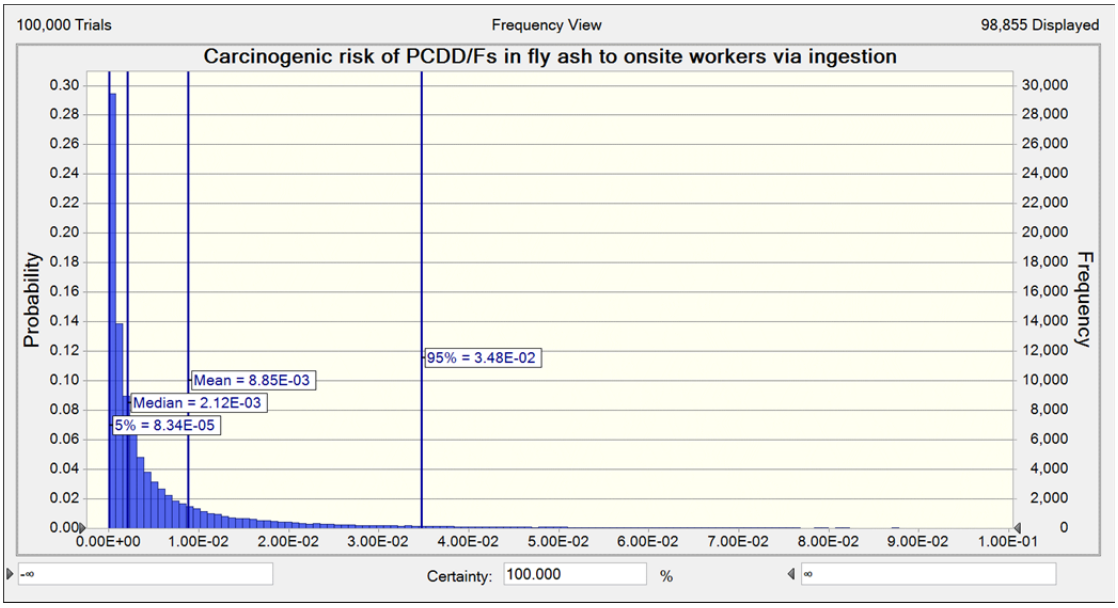


Fig. 5. The x-axis denotes the estimated carcinogenic risk. On the horizontal margins, the probability (left) and frequency (right) in the distribution are illustrated. (a) Predicted probability density distribution of total CR via three pathways for onsite workers. (b–d) Predicted probability density distribution of CR via different exposure pathways, respectively, inhalation, accidental ingestion, and dermal contact.

(c)



(d)

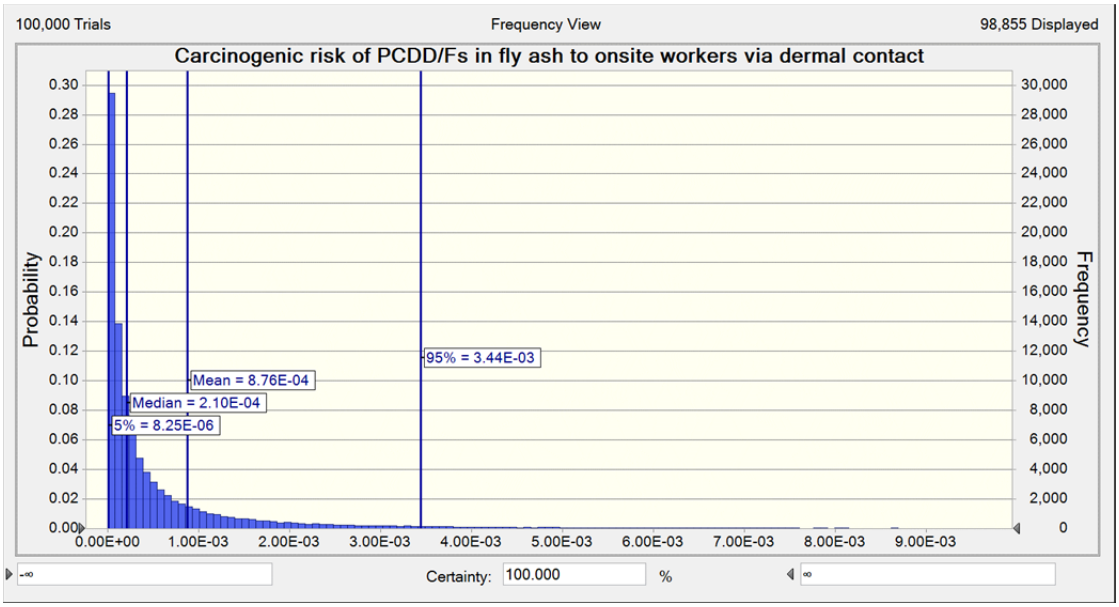


Fig. 5. (*continued*).

value was adopted for high-end estimates instead of the maximum value. The results demonstrated that the 95th percentile total CR value was 3.90×10^{-2}. Generally, carcinogenic risks between 10^{-6} and 10^{-4} are likely to cause cumulative health risk whereas carcinogenic risks greater than 10^{-4} suggest a high potential cancer risk, and those lower than 10^{-6} are considered negligible (Li *et al.*, 2014).

Thus, the results indicated that the detected 95th percentile total CR value was higher than the threshold value (10^{-4}), suggesting that there was a high degree of potential cancer risk for the onsite workers under consideration in this study.

In addition, different exposure pathways may contribute differently to carcinogenic risk. To determine the key exposure pathway, the CR for onsite workers via different exposure pathways were evaluated and depicted in Figs. 5(b)–5(d), respectively. The 95th percentiles carcinogenic risk via dermal contact and accidental ingestion were 3.48×10^{-3} and 3.55×10^{-2}, respectively, while the carcinogenic risk via inhalation was 2.84×10^{-9}, which was significantly lower than that through dermal contact and accidental ingestion. Clearly, accidental ingestion was the most important pathway for CR, followed by dermal contact and inhalation. Compared with the other two pathways, the carcinogenic risk caused by inhalation was almost negligible. This result is also consistent with earlier investigations on exposure to persistent organic pollutants (POPs) in fly ash and soils (Li *et al.*, 2016; Wu *et al.*, 2016). Moreover, residents near the plant had higher cancer risk related to PCDD/Fs than the general population, where pork, milk, and fish were considered as the major exposure pathways in Taiwan (Kao *et al.*, 2011; Tsai *et al.*, 2014). For the onsite workers, it is conceivable that the oral pathways such as ingestion of

water, food, and fly ash were the most dangerous routes associated with exposure to POPs. Hence, to protect the health of onsite workers in MSWI plants, they should be equipped with personal protection apparatus that prevents or reduces exposure to fly ash.

Non-Carcinogenic Risk Assessment

Through calculating the Hazard Quotient (HQ) and the Hazard Index (HI), the total non-carcinogenic risk of seventeen PCDD/Fs via three pathways was assessed for the onsite workers. The results are shown in Fig. 6. The total non-CR of PCDD/Fs varied from 1.67×10^{-4} to 2.99×10^{4}, with a median value of 26.6. Under most regulatory programs, a hazard index of more than one indicates that the exposed population is likely to experience adverse non-carcinogenic effects with the probability increasing with increases in the HI (Kienzler *et al.*, 2016). Here, the 95th percentile total non-CR value was 4.36×10^{2}, which was significantly higher than the threshold and thus considered to be causing a non-negligible hazard for the onsite workers. Subsequently, the non-CR values via different exposure pathways were estimated, as demonstrated in Figs. 6(b)–6(d). The 95th percentile value of non-CR via accidental ingestion was 3.82×10^{2}, whereas those via dermal contact and inhalation were 54.0 and 3.07×10^{-5}, respectively. Contributing more than 87.6% to the total non-CR, the

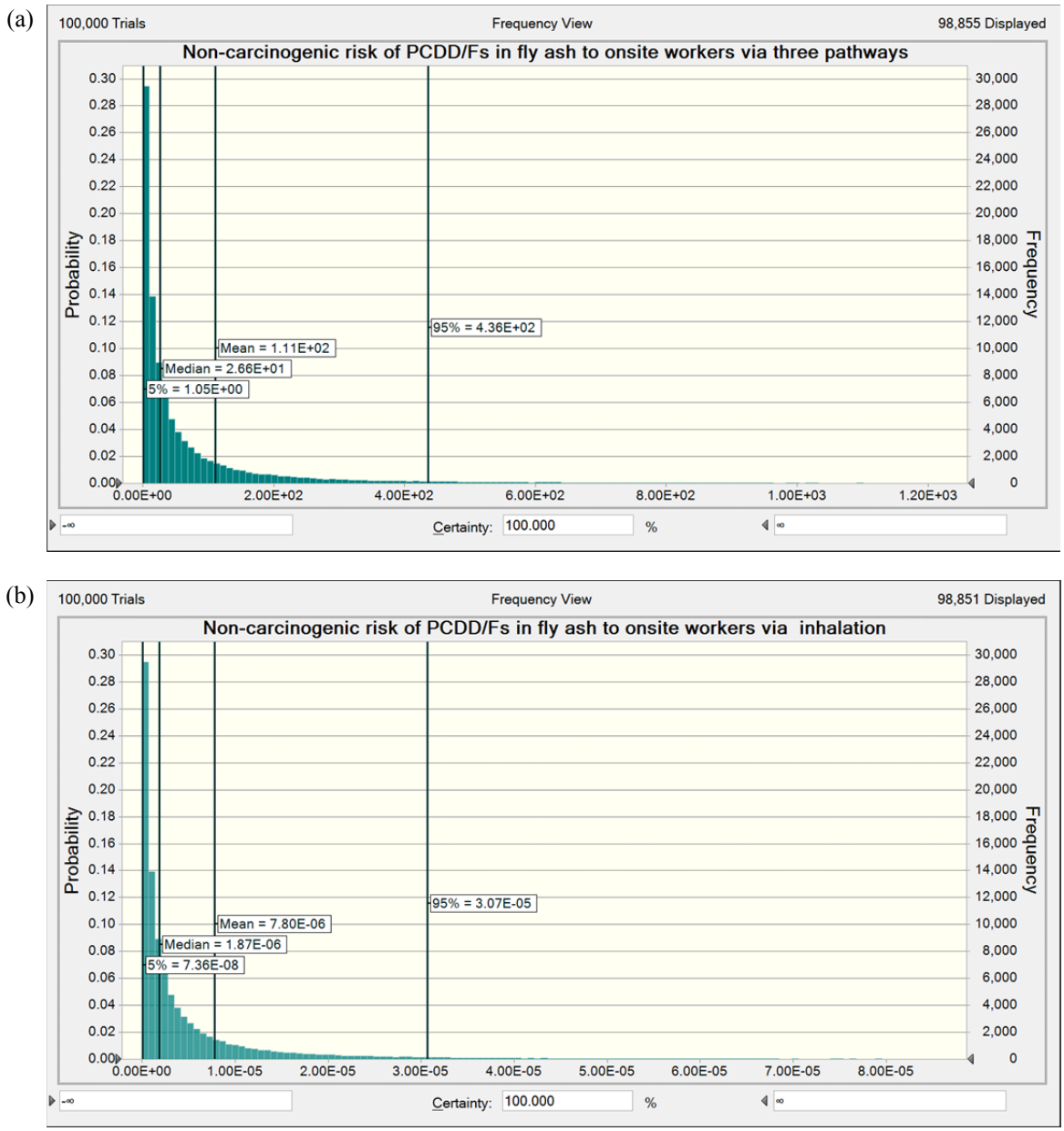
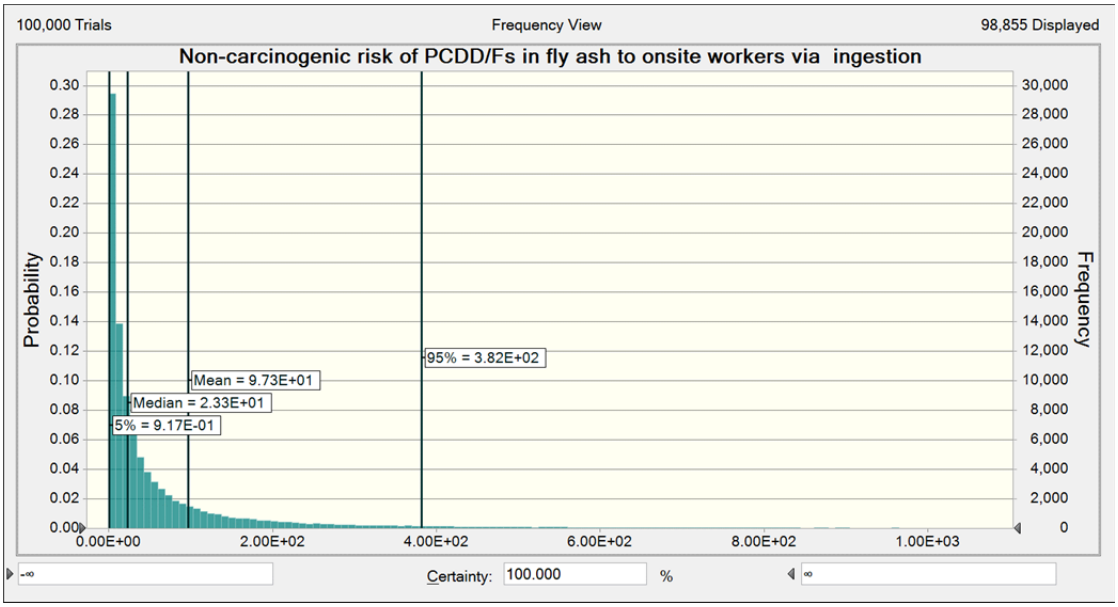


Fig. 6. The x-axis denotes the estimated non-carcinogenic risk. On the horizontal margins, the probability (left) and frequency (right) in the distribution are illustrated. (a) Predicted probability density distribution of total non-CR via three pathways for onsite workers. (b–d) Predicted probability density distribution of non-CR via different exposure pathways, respectively, inhalation, accidental ingestion, and dermal contact.

(c)



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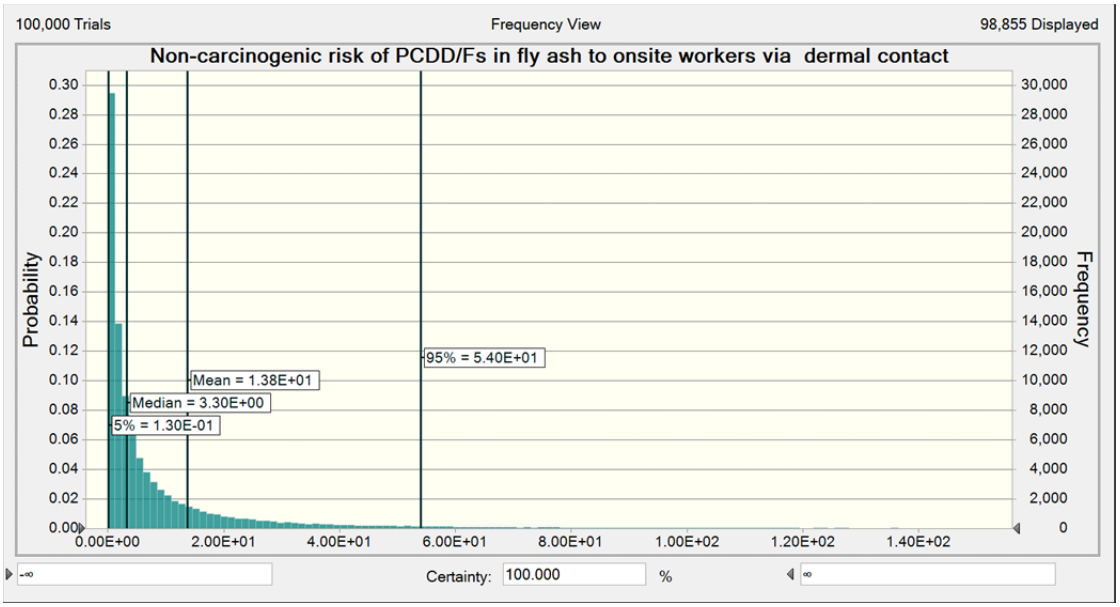

Fig. 6. (*continued*).

non-CR via accidental ingestion was significantly higher than that via the other two pathways. The ranking of the contribution to non-CR risk was accidental ingestion, dermal contact, and then inhalation, in that order. This finding is consistent with the results from previous reports (Li *et al.*, 2016; Wu *et al.*, 2016). Thus, the non-carcinogenic risk of PCDD/Fs in fly ash should not be neglected.

Also, considering that the pathway of accidental ingestion was a pivotal contributor to both non-CR and CR, specific protective measures should be provided for onsite workers to reduce health risks via accidental ingestion.

Sensitivity Analysis of Parameters in the Monte Carlo Simulation

Sensitivity analysis a useful approach by which to identify the most sensitive parameters in a model and to quantify how parameter uncertainties influence the outcomes (Saltelli, 2002; Alam *et al.*, 2016). Here, the results of the sensitivity analysis for the CR and non-CR assessments are shown in Fig. S1. The results suggest that Concentration (CC) and ED were the most sensitive parameters in both the CR and non-CR assessments. The sum contributions of CC and ED accounted for more than 99% of the total variance in the health risk assessment, in which concentration was found to be the most influential parameter. These results were consistent with the findings of Yang *et al.* (2015). Although some previous studies have revealed that ED is the dominant factor, they still found that CC and ED were the two most influential factors in risk assessments (Li *et al.*, 2016; Tong *et al.*, 2018). Thus, on one hand, to improve the assessment accuracy of health risks due to PCDD/Fs, a more accurate representation of the probability

distribution for CC and ED might be helpful. On the other hand, controlling CC and ED would be an effective method by which to improve existing risk mitigation strategies.

Other parameters, such as BW and EF, totally contributed less than 1%. Similar results have also been reported (Yang *et al.*, 2015; Li *et al.*, 2016). Although the evaluated contribution of these parameters seems negligible, they probably disclosed the limitations/shortcomings of the current health risk assessment models and contributed substantially to the total uncertainties. Firstly, in current risk prediction model, the specific division of labor and professional differences was not considered. Luo *et al.* (2014) unveiled the great difference in health risk between demolition workers and general industrial workers. It is likely that in the MSWI plant, there may be different health risks between workers responsible for recycling and disposal of fly ash and workers in many other occupations, including management and human resources. Secondly, CC is the sum of converted concentrations for 17 PCDD/Fs in fly ash based on toxic equivalents (TEFs). The sensitivity analysis revealed that CC is the most sensitive factor and has a significant influence on risk evaluation. Cumulative and synergistic effects on human health have been revealed by analysis of combinations of different food additives or drugs (Park *et al.*, 2009; Foucquier and Guedj, 2015). Combinational effects of two or more PCDD/Fs were neglected and may have created uncertainties in the present model. In addition, some parameters (such as BW and SA) were directly retrieved from the USEPA (U.S. Environmental Protection Agency), which might not be applicable to all races. What's worse is that some parameters were obtained based on animal (such as beagle dogs) experiments. The unreliability of applying animal experimental results to human biology and diseases is increasingly being recognized (Akhtar, 2015). Extrapolating animal test results to humans may result in unique uncertainties. Hence, it warrants further investigations to develop more direct, human-based technologies and to build more accurate statistical models for health risk assessments related to POPs.

CONCLUSIONS

In the present study, PCDD/Fs in fly ash from an MSWI in Southern Taiwan was analyzed from several perspectives, mainly including concentration distribution, TEQ and characteristic indexes, clustering and correlation analyses, carcinogenic and non-carcinogenic risk assessments, and sensitivity analyses. The results can be summarized as follows: (1) Through a multiple comparison analysis, it was revealed that four types of congeners (OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF) have significantly higher concentrations than other species ($p < 0.01$). (2) 2,3,4,7,8-PeCDF was the main contributor to the total toxic equivalent concentration (TEQ); and the top three candidate indicators of total TEQ are OCDF, 1,2,3,4,6,7,8-HpCDF, and 2,3,4,7,8-PeCDF, in which OCDF might be most powerful indicator in fly ash from similar sources. (3) It was indicated that all congeners correlated positively with each other. All the average R^2 values across congeners were in the range of 0.707-0.939, with minimum and maximum R^2 values on 1,2,3,7,8,9-HxCDF, and 2,3,4,6,7,8-HxCDF, respectively. Meanwhile, the results of the cluster analysis revealed the specific features of several congeners (such as 1,2,3,7,8,9-HxCDF, OCDD, and OCDF). In addition, most samples from the winter season were grouped into one cluster. This implied that cluster analysis can provide the finger characteristics of PCDD/Fs in fly ash and can be a useful index for different seasons. (4) Through assessing health risk with a Monte Carlo simulation, both the 95th percentile carcinogenic risk and non-carcinogenic risk for onsite workers was shown to exceed the threshold limit and should be considered as significant risks for onsite workers. Further, it was found that the pathway of accidental ingestion was a pivotal contributor to both non-CR and CR. Therefore, specific protective measures should be provided for onsite workers to reduce health risks via accidental ingestion (5) The results of the sensitivity analysis suggested that concentration (CC) and exposure duration (ED) are the most two sensitive parameters according to both the CR and non-CR assessments. Overall, the findings in this study could be useful for development and implementation of health risk strategies for onsite workers in MSWI plants.

ACKNOWLEDGMENTS

This research was supported by the Fundamental Research Funds for the Central Universities (Grant No. JZ2017YYPY0899).

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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Received for review, December 20, 2017
Revised, February 4, 2018
Accepted, February 4, 2018