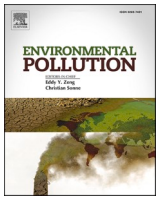
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Characteristics, correlations and health risks of PCDD/Fs and heavy metals in surface soil near municipal solid waste incineration plants in Southwest China☆

Xin Bo [a], Jing Guo [b], Ruxing Wan [c], Yuling Jia [d], Zhaoxu Yang [e], Yong Lu [f], Min Wei [g,*]

[a] Department of Environmental Science and Engineering, Beijing University of Chemical Technology, Beijing, 100029, China
[b] School of Economics and Management, Beihang University, Beijing, 100191, China
[c] School of Economics and Management, Beijing University of Chemical Technology, Beijing, 100029, China
[d] Sichuan Environment and Engineering Appraisal Center, Chengdu, 610041, China
[e] Zhongke Sanqing Technology Co., Ltd., Beijing, 100020, China
[f] Pony Testing International Group Co., Ltd., Beijing, 100080, China
[g] College of Geography and Environment, Shandong Normal University, Ji'nan, 250014, China

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ABSTRACT

As primary anthropogenic emission source of toxic pollutants such as heavy metals and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), municipal solid waste (MSW) incineration has caused worldwide concern. However, a comprehensive analysis of the pollution characteristics and health risks of PCDD/Fs and heavy metals in soils around MSW incineration plants is lacking. In this study, 17 PCDD/Fs and 11 heavy metals in soil samples collected near MSW incineration plants in Sichuan province were investigated to evaluate their pollution characteristics and potential health risk. Sichuan was selected as the study area because the MSW incineration amount in this province ranks first among all inland provinces in China. The PCDD/Fs concentrations ranged from 0.30 to 7.50 ng I-TEQ/kg, which were significantly below risk screening and intervention thresholds. Regarding heavy metals, principal component analysis suggested that Hg, Pb and Zn were the primary metals emitted from the MSW incineration plants. Cluster analysis of PCDD/Fs and heavy metals showed that of PCDD/Fs homologs and heavy metals (e.g., Hg, Pb, Zn and Cd) were clustered into one group, indicating the coexistence and coaccumulation of heavy metals (especially Hg, Pb, Zn, and Cd) and PCDD/Fs in soil. These heavy metals are thus candidate tracers for PCDD/Fs in soil near MSW incineration plants. A health risk analysis found that the carcinogenic and non-carcinogenic risks of PCDD/Fs and heavy metals (except for Ni) in the soil samples were all within acceptable levels. This study provides new insights into correlations and health risks of PCDD/Fs and heavy metals in surface soil near MSW incineration plants. The findings have implications for future studies of environmental and human health risk analysis related to waste incineration.

1. Introduction

Over the past 40 years, with rapid urbanization and a high population growth rate, the amount of municipal solid waste (MSW) produced in China has been steadily increasing, from 31.3 million tons in 1980 to 242.1 million tons in 2019, at an annual growth rate of 5.39% (MOHURD, 2020). The growth rate of MSW has been significantly more rapid than that of the Chinese population (0.90%). Today, there are three main disposal methods for MSW, landfilling, composting and incineration (Tian et al., 2012). A lack of landfill and composting sites has caused incineration to become the most common alternative for disposing MSW in China. During incineration, solid waste is converted into residue and gaseous substances, which significantly reduces the volume and weight of the waste by 90% and 70%, respectively (Tian et al., 2012). Therefore, incineration has widely been accepted as an effective method for reducing waste and the number of MSW incineration plants in China is steadily increasing (Liu et al., 2012; MOHURD, 2020).

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* Corresponding author.
E-mail address: minwei@sdnu.edu.cn (M. Wei).

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Incineration is controversial because it emits potentially harmful compounds associated with human health risks. Previous studies have indicated that toxic chemicals such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and heavy metals can be released during MSW incineration and be deposited in soil via wet and dry deposition, posing a serious threat to human health because they do not readily biodegrade and have a long biological half-life (Li et al., 2018). Concerns about soil contamination and health risks have recently caused public concern in China, particularly regarding PCDD/Fs and heavy metals derived from MSW incineration plants (Li et al., 2019b; Wang et al., 2020a; Zhong et al., 2020). Therefore, it is essential to better understand the concentrations and pollution characteristics of toxic compounds in soil near waste incinerators and to assess their potential health risks to nearby populations.

Previous studies have investigated many toxic pollutants in soil around MSW incineration plants (recent works are listed in Table 1). However, few studies have comprehensively explored the characteristics and health risks of PCDD/Fs and heavy metals (Rovira et al., 2018), and little research has focused on the relationships between PCDD/Fs and heavy metals in the soil around MSW incineration plants. In addition, identifying PCDD/Fs in soil is challenging owing to high testing costs, advanced technical requirements, numerous homologs, and the typically low concentrations of PCDD/Fs in soil (Eljarrat and Barcelo, 2002; Peng et al., 2020). In this study, Sichuan province was investigated because its waste incineration amount ranked first among all inland provinces in 2019 (Table S1). We analyzed soil samples collected near incinerators for the presence of PCDD/Fs and heavy metals and explored their correlation. We also quantitatively evaluated their health risks (including carcinogenic risk (CR) and non-carcinogenic risk (NCR)). The findings of this study provide new ideas for the further rapid identification and detection of PCDD/Fs.

2. Materials and methods

2.1. Study area

Sichuan province is situated in Southwest China (26°03′–34°19′ N, 97°21′–108°12′ E) and covers an area of 486,000 km^2. Sichuan has a subtropical monsoon climate with a mean annual precipitation of 1000–1200 mm and a mean annual temperature of 16–18 °C. In this study, soil samples were collected from six waste incineration power plants that had been operating for at least 4 years, with annual disposal capacities of 2–8 million tons. The detailed operating parameters of the MSW incineration plants are shown in Table S2. The combustion systems of the plants utilize a grate firing incinerator. The National Energy Administration has proposed that the flue gas treatment system of waste incineration power generation plants should include a deacidification system, an activated carbon system, a dust removal system, and a desalination system (DL/T 1967-2019). The flue gas treatment systems of the six waste incineration plants in this study all met this requirement.

2.2. Sample collection

Owing to the influence of topography and meteorological conditions, the diffusion of pollutants around the plant areas varied greatly, resulting in varied spatial distribution patterns. It is impossible to precisely predict the concentrations and distribution patterns of pollutants in the atmosphere and soil during wet and dry deposition. In particular, specific regions of high atmospheric and soil settlement concentrations cannot be determined. Therefore, the CALMET meteorological model was used to simulate a regional three-dimensional meteorological field, which was then input to CALPUFF for simulation and prediction to determine the areas with high atmospheric and soil deposition concentrations. Finally, a total of 16 soil sampling locations (S1–S16) with high predicted concentrations near the six waste incineration plants were selected. The sampling locations included a control group (S3, S6,

Table 1
Recent studies on PCDD/Fs and heavy metals in soil near MSW incineration plants.

	Heavy metal (mg/kg)																		PCDD/Fs	Total	Health risks	
	Fe	Sb	Mn	As	Ni	Co	Cr	Cu	Hg	Cd	Zn	Pb	Tl	V	Sn	Ag	Li	Ti			CR	NCR
This study	×	1.0	672.6	6.9	40.5	15.8	81.2	34.1	0.2	0.3	193.6	25.7	×	×	×	×	×	×	1.50	**11 + 1**	✓	✓
Deng et al. (2016)	×	×	×	×	×	×	×	×	29.7	×	×	×	×	×	×	×	×	×	×	**1**	×	×
Han et al. (2016)	×	×	546	7.7	24	×	52	28	0.09	×	100	19	×	×	×	×	×	×	×	8	✓	✓
Meng et al. (2016)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	2.0	1	×	×
Domingo et al. (2017)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	1.7	1	✓	✓
Li et al. (2017)	×	×	321.5	×	×	×	37.1	×	×	0.3	×	58.8	×	×	×	×	×	×	×	4	✓	✓
Li et al. (2018)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	10.8	1	✓	✓
Ma et al. (2018)	×	×	×	31.2	31.8	×	76.4	30.5	0.14	0.4	120.2	26.7	×	×	×	×	×	×	×	8	✓	✓
Ngo et al. (2018)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	10.4	1	×	×
Rovira et al. (2018)	×	0.24	330	4.7	11.1	4.9	8.4	23	0.01	0.18	×	17.2	0.08	17.1	1.3	×	×	×	0.38	13 + 1	✓	✓
Li et al. (2019a)	×	×	613.2	16	30.2	×	84.9	50.8	0.05	0.67	177	53.4	×	×	×	×	×	3161	×	11	✓	✓
Li et al. (2019b)	×	1.2	×	×	29.7	×	77.2	31.4	0.12	0.22	107.2	32.2	×	×	×	×	×	×	×	8	×	×
Xu et al. (2019)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	1.43	1	✓	✓
Zhong et al. (2019)	493358	×	856.2	×	×	×	249	34.6	<0.01	<0.01	80.8	37.2	×	×	×	×	×	×	×	8	×	×
Deng et al. (2020)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	1.79	1	✓	✓
Nadal et al. (2020)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	0.23	1	×	×
Van Drooge et al. (2021)	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×	6.26	1	×	×

Note: PCDD/Fs (ng I-TEQ/kg). I-TEQ = International toxic equivalency.

S10, S13, and S16), maximum landing concentration locations (S2, S5, S9, S12, and S15), and maximum subsidence concentration locations (S1, S4, S7, S8, S4, S11, and S14) (Fig. 1). Detailed information on the sampling locations is shown in Table S3. Considering the short operating time of Plant C and the relatively low soil accumulation of pollutants according to the settlement results predicted by the CALPUFF model, only one sample was collected near Plant C. The selection of sampling locations is critical to accurately assess pollutant levels. Uncertainty may arise from human activities (e.g., straw burning, herbicide use, and vehicle transportation) in the vicinity of sampling or from excessively low PCDD/F and heavy metal concentrations in the soil. Therefore, soil background samples in areas not obviously impacted by human activities were also collected.

The surface soil samples were obtained by mixing five different specimens whereby one specimen was collected from a center point and the others were collected in each of the four directions 5 m from the center. Samples for PCDD/Fs and heavy metal analysis were collected in glass bottles and polyethylene containers, respectively, and sealed to prevent evaporation and contact with the outside environment.

2.3. Sample treatment and analysis

The detailed soil analysis procedures included sample extraction, clean-up, and the determination of PCDD/Fs and heavy metals concentrations (Li et al., 2010; Zhou et al., 2019). The PCDD/Fs, analysis was carried out in accordance with HJ 77.4–2008 (MEPC, 2008), and all solvents used in the extraction and clean-up procedures were of pesticide residue analysis-grade, acquired from CNW (Shanghai, China). For extraction, the soil samples were dried at room temperature, and sieved through a 60-mesh sieve, and extracted via the selective pressurized liquid extraction method in an ASE 300 system (Dionex, Sunnyvale, CA, USA) (Subedi et al., 2015). In the clean-up process, the extracts were changed to n-hexane solvents and transferred to a multilayer silica gel column(Merck, Germany). The PCDD/Fs were eluted with 150 mL of hexane and then flowed over a silica-gel dispersed carbon column for further PCDD/Fs fractionation. The PCDD/Fs were eluted with 40 mL of toluene. All eluted fractions were concentrated to 20 μL and spiked with 5 μL of ^{13}C-labeled standards (Wellington, Ontario, Canada). For PCDD/Fs determination, the toxic 2,3,7,8-substituted PCDD/Fs were quantitatively measured by high resolution gas chromatography/high resolution ass spectrometry (HRGC-HRMS) (ThermoFisher Trace1310, DFS), and the PCDD/Fs were analyzed using a DB-5MS column (60 m × 0.25 mm i.d. × 0.25 μm). Highly purified helium was used as the carrier gas at a constant flow rate of 1.0 mL/min. The temperature program of the capillary column was as follows: (1) initial oven temperature of 140 °C, held for 1 min; (2) increased at 20 °C/min to 200 °C, held for 1 min; (3) increased at 5 °C/min to 235 °C, held for 16 min; (4) increased at 5 °C/min to 235 °C, held for and 7 min, respectively; (4) increased at 5 °C/min to 310 °C, held for 10 min. The temperatures of the injector, interface, and ion source were set at 270 °C, 280 °C and 280 °C, respectively. The HRMS was operated in electron impact (45 eV) and selected ion monitoring mode at a resolution of R > 10000 (10% valley). A method blank included in each batch of samples to confirm the absence of interferences. The analysis method used in this study was validated by the recoveries of ^{13}C-labeled internal quantification standards. The method detection limits of the instrument, method, and samples were confirmed regularly. In this study, the sample detection limit of 2,3,7,8-TCDD was 0.009 ng/kg based on a sampling weight of 10 g. The total toxic equivalency of the 17 2,3,7,8-chlorine substituted PCDD/F congeners was calculated by multiplying the measured concentration of each congener by the toxicity equivalent factor (GB 36600-2018).

The soil samples were analyzed for heavy metals in accordance with methods GB/T 22105.1-2008 (for Hg and As, using atomic fluorescence spectrometry), GB/T 17141-1997 (for Pb and Cd, using graphite furnace atomic absorption spectrophotometry), HJ 491-2019 (for Cu, Ni, Zn and Cr, using flame atomic absorption spectrophotometry), HJ 680-2013 (for Sb, using microwave dissolution/atomic fluorescence spectrometry), and EPA3050B:1996 and EPA6010D:2008 (for Al, Co, Fe and Mn, using acid digestion inductively coupled plasma atomic emission spectrometry). All of the methods met the Testing Authority Certification requirements. Each soil sample was dried in an oven at 105 °C, sieved through 100-mesh sieve, digested with 8 mL of HNO_3:HF:H_2O_2 (v/v) at a 5:2:1 ratio in a microwave digestion system (MARS 5, CEM, Matthews, NC, USA), and incubated at 165 °C for 20 min. The concentrations of heavy metals (including Fe, Al, Sb, Mn, As, Ni, Co, Cr, Cu, Hg, Cd, Zn, and Pb) were measured by inductively coupled plasma mass spectrometry (Thermo Scientific iCAP Qs ICP-MS, USA).

For quality assurance and quality control, program blank and matrix spiked samples were processed and analyzed together with the filed samples and used to verify the concentration data. The analysis of the program blank showed that the system and glassware did not affect the concentration determinations. Two parallel aliquots from all samples were randomly selected for repeatability testing. The signal-to-noise ratio of the 17 PCDD/F congeners in the blank sample was 3:1. This was used to estimate the HRGC-HRMS detection limits of the congers, which ranged from 0.001 to 0.005 ng/g. The congener recoveries were between 28% and 124%. In the heavy metals analysis, a national standard soil sample (GBW07428, GBW07604; Center for Certified Reference Materials, China) was included in each set of samples to validate the analytical procedures. The recovery rates of the national standard soil sample were 103.28–114.48% for Hg, 96.84–99.37% for As, 90.76–92.12% for Cr, 101.45–107.97% for Ni, 86.64–90.95% for Cu, 97.80–109.62% for Zn, 107.78–113.33% for Cd, 94.86–100.34% for Pb, 107.78–113.33% for Cd, 97.66–100.78% for Co, 94.76–95.61% for Mn, and 137.63–159.14% for Sb, respectively.

2.4. Contamination assessment

The contamination risk of the PCDD/Fs and heavy metals in the soil were evaluated using different methods. For PCDD/Fs, potential environmental effects were evaluated the as international toxic equivalency (I-TEQ) of PCDD/Fs, which was calculated using the toxic equivalency factor in accordance with the International Toxic Equivalent System (NATO/CCMS, 1988). For heavy metals, two assessment methods were used to quantify the pollution level: i.e., the single pollutant index (Wang et al., 2020b) and Nemerow pollution index (Martínez-Guijarro et al., 2019). Three indexes evaluated the contamination risk of heavy metals by comparing the metal in the soil samples with the control group from the national soil environmental quality standard value. The single pollutant index is calculated on the basis of the soil pollution classification standard as follows (Li et al., 2016; Wang et al., 2020b):

$$P_i = \frac{C_i}{S_i}, \quad (1)$$

where P_i represents the single pollutant index of metal i in the soil, C_i is the average concentration of metal i in the samples, and S_i is the soil environmental quality reference value of metal i. The background values were obtained from the Ministry of Environmental Protection China (MEPC, 1990).

On the basis of the single pollutant index, the degree of soil contamination can be classified into four levels, as shown in Table S4. $P_i \leq 1$ indicates that the soil has not been contaminated by metal i; $1 < P_i \leq 2$ means that the soil is polluted to a slight-degree; $2 < P_i \leq 3$ indicates that the soil is moderately contaminated by metal element i; $P_i > 3$ indicates that the soil is contaminated to a strong-degree.

However, this method calculates an index for only one element in soil a time and does not clarify the comprehensive contamination status. Therefore, the Nemerow comprehensive index (N_{ci}) method was used to reflect the degree of contamination by multiple heavy metals to investigate their impact on soil quality (Li et al., 2014a). In the present study,

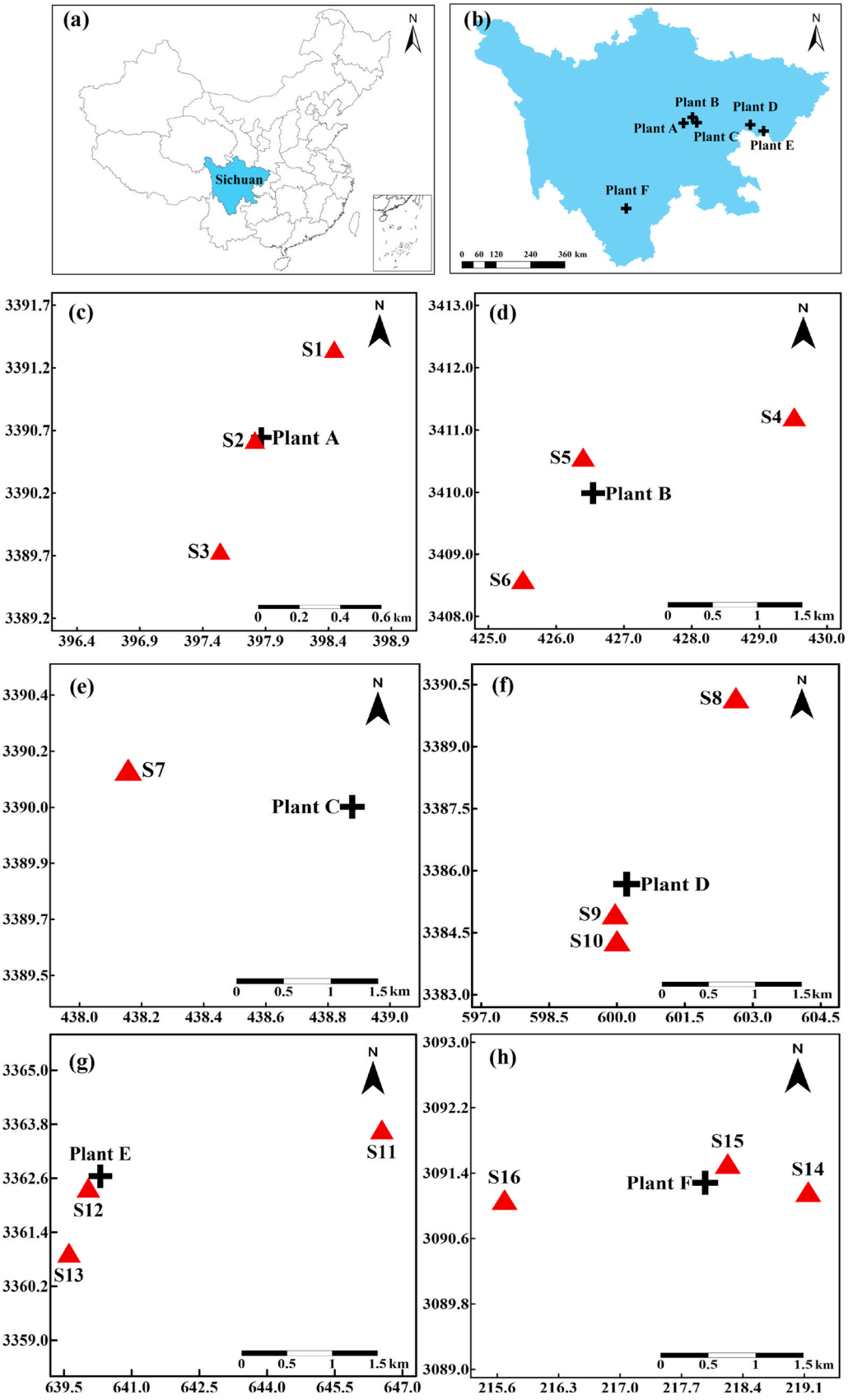


Fig. 1. Location of the study area and sampling sites (UTM coordinates, Unit: km). **a**, the study area. **b**, the location of six MSW incineration plants. **c-h**, the distribution of sampling sites.

the N_{ci} was applied as the primary evaluation method and was supported by the single pollutant index values. The N_{ci} is calculated as follows:

$$N_{ci} = \sqrt{\frac{\left(\overline{P}_i^j\right)^2 + \left(P_{i\max}^j\right)^2}{2}}, \quad (2)$$

where $\overline{P}_i^j$ represents the average value of P_i at sample location j, and $P_{i\max}^j$ is the maximum value of P_i at sample location j. According to the N_{ci} method, the degree of soil contamination can be divided into five levels, as shown in Table S5.

2.5. Health risk assessment

Human health risks include *CR* and *NCR* from exposure to trace elements, as assessed by the USEPA standard model. The main environmental exposure pathways associated with soil contaminant chronic daily intake (*CDI*) are dust ingestion (CDI_{ing}), air inhalation (CDI_{inh}), and dermal contact (CDI_{der}). On the basis of behavioral and physiological differences, residents near the sampling areas were categorized into three age groups (children, teens and adults) for PCDD/Fs risk analysis and into three age groups (children, males and females) for heavy metals risk analysis. For soil exposure, the *CDIs* of pollutants were calculated through different intake methodologies. Formulas (3), (4), and (5) were used to calculate the PCDD/Fs *CDIs* for children, teenagers, and adults and heavy metals *CDIs* of children, males, and females, as follows:

$$CDI_{ing} = \frac{C_s \times IngR \times EF \times ED}{AT \times BW} \times 10^{-6}, \quad (3)$$

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

$$CDI_{inh} = \frac{C_s \times InhR \times EF \times ED}{PEF \times AT \times BW}, \quad (5)$$

where CDI_{ing}, CDI_{inh} and CDI_{der} are the average daily doses through ingestion, inhalation of soil particles and dermal contact, respectively in mg/(kg•day). C_s is the concentration of pollutants in the soil ((PCDD/Fs (mg I-TEQ kg^{-1}); heavy metals (mg kg^{-1})). *IngR* is the soil ingestion rate (mg/day). *InhR* is the air inhalation rate (m^3/day). *PEF* is the particulate emission factor (m^3/kg). *SA* is the exposed surface area of the skin (m^2). *AF* is the skin adherence factor (mg/(cm^2 •day)). *ABS* is the dermal absorption factor (unitless). *EF* is the annual exposure frequency (day/year). *ED* is the exposure duration (years). *AT* is the average exposure time (days). For carcinogen risk, the *AT* of the three age groups was 70 × 365 days. For non-carcinogen risk, the *AT* values of PCDD/Fs for children, teenagers, and adults were 11 × 365, 6 × 365, and 53 × 365 days, respectively. *BW* is the body weight (kg). The values, parameters distributions, and health risk assessment references used in this study are listed in Table S6–S8.

The *CR* from PCDD/Fs and heavy metals in the soil for the three age groups was calculated by formula (6) (Li et al., 2018):

$$CR_{soil} = \sum_{i=1}^{3} CDI_i \times SF_i, \quad (6)$$

where *CR* represents the total carcinogenic risk through the three exposure pathways (unitless); *SF* is for the carcinogenicity slope factor; and *i* indicates the three exposure pathways.

The *NCR* from soil pollutants was evaluated using the Hazard Index (*HI*), calculated according to formula (7):

$$HI_{soil} = \sum_{i=1}^{3} \frac{CDI_i}{RfD_i} \quad (7)$$

where *HI* indicates the total non-carcinogenic risks (unitless), and *RfD* is the reference dose concentration for the three exposure pathways (mg/(kg•day)).

Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were employed to investigate the similarities and differences within the pollutant patterns in the soil samples, to reduce the data dimensionality for further analysis (Zhao et al., 2008). All calculations, analyses, and visualizations were executed in RStudio, v.4.0.2.

3. Results and discussion

3.1. PCDD/Fs concentration in soil

The concentrations and I-TEQ values of PCDD/Fs in the soil samples collected near the target studied MSW incineration plants are shown in Table S9. The PCDD/Fs concentrations and I-TEQ values varied from 7.14 to 858.09 ng/kg and from 0.25 to 7.50 ng I-TEQ/kg, respectively, with average values of 97.48 ng/kg and 1.54 ng I-TEQ/kg. The I-TEQ values of the PCDD/Fs were lower than the risk screening (40 ng I-TEQ/kg) and risk intervention values (400 ng I-TEQ/kg) obtained from the risk control standards for soil contamination of development land (GB 36600-2018).

The homolog profiles of the PCDD/Fs at each sampling locations are shown in Fig. 2. The distribution of the homolog profiles varied greatly among the soil samples (Table S10). Generally, the high-chlorinated PCDD/Fs (i.e., PCDDs) were a major component of the homolog profiles from samples near MSW combustion sources (representing 91.14% of the total PCDD/Fs concentration). By contrast, low-chlorinated PCDD/Fs (i.e., PCDFs) represented 8.86% of the concentration, which was consistent with the results of previous studies (Floret et al., 2007; Oh et al., 2006; Xu et al., 2009). Regarding the PCDD class, the most abundant chlorinated PCDD/Fs was 2,3,7,8-TeCDD, accounting for 10.17% of the total I-TEQ content, indicating that this compound greatly affects by the level of organochlorine pollution. Regarding the PCDF class, 2,3,4,7,8-PeCDF was the predominant component among the PCDFs homologs and accounted for 46.18% of the total I-TEQ content.

The PCDFs/PCDDs (F/D) ratio can effectively indicate the source of PCDD/Fs in environmental media. If the F/D ratio is greater than 1, the PCDD/Fs comes from combustion sources. By contrast, an F/D ratio of less than 1 indicates that there are multiple sources of PCDD/Fs in the atmosphere, among which waste incineration may be a key source (Li et al., 2018). The F/D ratio in the soil samples ranged from 0.23 to 5.41, with an average of 1.46. Among the 16 soil samples, the F/D ratios of six were less than 1, suggesting that combustion was not the main source of PCDD/Fs in the soil, and the investigated MSW incineration plants may have had a limited effect on the PCDD/Fs concentrations (Li et al., 2018). The F/D ratios of the other 10 samples were greater than 1, which further illustrated that PCDFs were the main homolog profiles affecting the total I-TEQ content of PCDD/Fs in the soil samples (Li et al., 2018). The average PCDD/Fs soil concentrations near the six plants ranged from 0.25 to 5.4 ng I-TEQ/kg, with average of 1.39 ng I-TEQ/kg (Fig. 2). The average PCDD/Fs soil concentrations of the six plants were in the order of Plant A > Plant B > Plant E > Plant D > Plant F > Plant C, which may be associated with their durations of operation ($r = 0.76$, $p < 0.1$, 95% confidence interval (CI) = [−0.14, 0.97]). The results showed an increasing trend between the operating time of the plant and the accumulation of pollutants in the surrounding soil. Such trends have been found in previous studies (Xu et al., 2009; Lin et al., 2020).

The I-TEQ values of PCDD/Fs in soil collected near different anthropogenic emission sources are summarized in Table S11. The I-TEQ values of PCDD/Fs in this study were similar to or lower than those found near other waste incinerators in China, such as those near Beijing (Li et al., 2014b), Hangzhou (Jin et al., 2012) and Shanghai (Deng et al., 2011). However, they were higher than those found in Spain (Vilavert et al., 2012; Vilavert et al., 2014; Rovira et al., 2014) and Italy (Colombo et al., 2013). Therefore, while the soil contamination levels found near the investigated MSW incineration plants in Sichuan were relatively low, additional efforts are needed to further limit PCDD/Fs soil

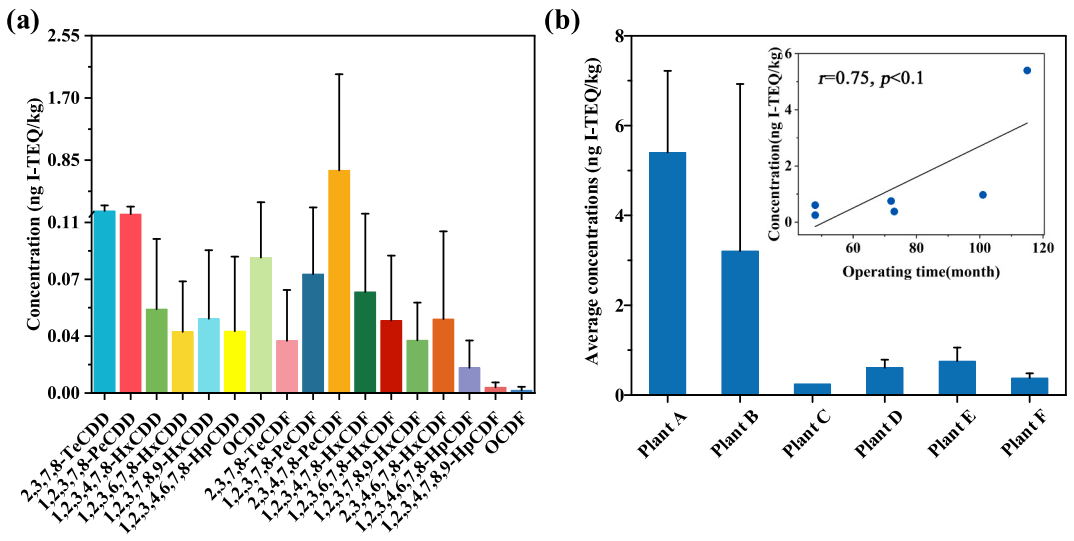


Fig. 2. Homolog profiles and average concentrations of PCDD/Fs. **a**, Homolog profiles of PCDD/Fs for all samples. **b**, Average soil concentrations near the of six plants.

contamination. Compared with other emission sources, the I-TEQ levels found in this work were significantly lower than those found near electronic waste (EW) disposal plants (Xu et al., 2013), EW acid leaching sites (Leung et al., 2007), EW recycling plants (Chakraborty et al., 2018), and iron and steel plants (Sun et al., 2017), but they were close to those of cement plants (Schuhmacher et al., 2009). In summary, the PCDD/Fs soil concentration generated by the MSW incineration plants was found to be lower than that generated by other anthropogenic emission sources in China, especially when compared with EW emission sources.

3.2. Heavy metal concentrations in soil

Heavy metal contamination in soil near the MSW incineration plants was investigated. The concentrations of the 11 investigated heavy metals are listed in Table S12, and the results of the statistical analysis are shown in Table S13. The mean heavy metal concentrations were in the following order: Hg < Cd < Sb < As < Co < Pb < Cu < Ni < Cr < Zn < Mn. As single pollutants, Mn and Zn were the predominant heavy metals, with concentrations of 672.56 and 193.58 mg/kg, respectively. In contrast, Hg, Cd, and Sb had the lowest mean concentrations (0.15, 0.32 and 0.96 mg/kg, respectively). All metal concentrations in the collected soil samples were lower than the limits set by the risk control standard for soil contamination of agricultural land (GB 15618-2018). Regarding the single pollutant index, only As and Sb had the lowest degree of pollution (uncontaminated). The highest index values were for Cd and Hg (high contamination), followed by Zn (moderate contamination), and Pb, Cu, Cr, Co, Ni and Mn (low contamination). Based on Nemerow's comprehensive index (N_{ci}), most sampling locations (S1–S5, S9–S10, and S12–S16) had a high level of contamination (N_{ci} >3). The other sample locations S6–S8 and S11) had a moderate level of contamination (2< N_{ci}≤3). These results indicated that the soils around the MSW incineration plants had differing amounts of heavy metal contamination. Soil near Plant A had the highest levels of heavy metal (e.g., Cd, Hg, Pb, Zn and Cu), which may have been from the coating and painting activities of three large furniture factories around Plant A; these activities are known emission sources of Cd, Cu, and Pb (Tong and Lam, 2000). The highest heavy metal concentrations of Plant B, C, and D were Cr; As and Sb; and Ni, Mn, and Co; respectively, as shown in Fig. 3.

Fig. 4 compares the heavy metal concentrations found in soil near the MSW incineration plants with those found in previous studies. The

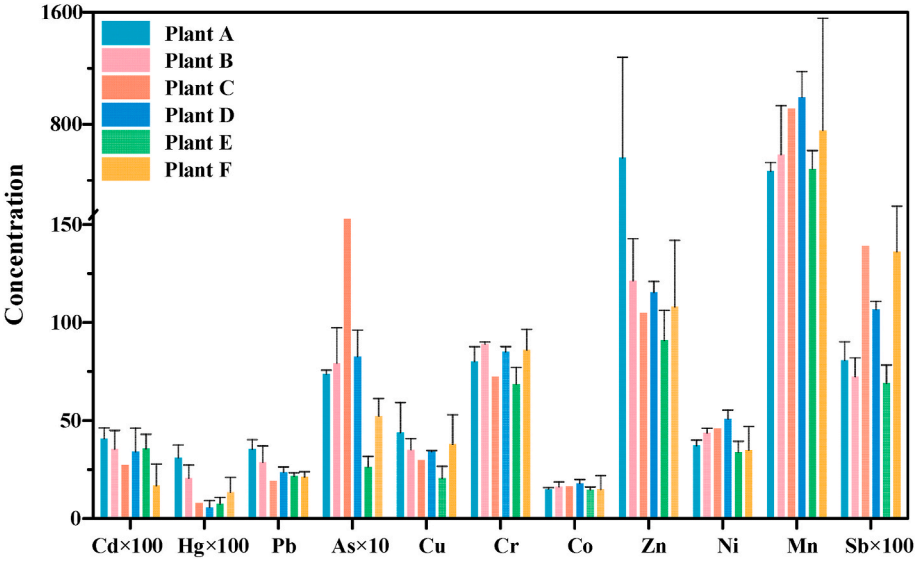


Fig. 3. Heavy metal concentrations (mg/kg) in surface soil near the studied MSW incineration plants.

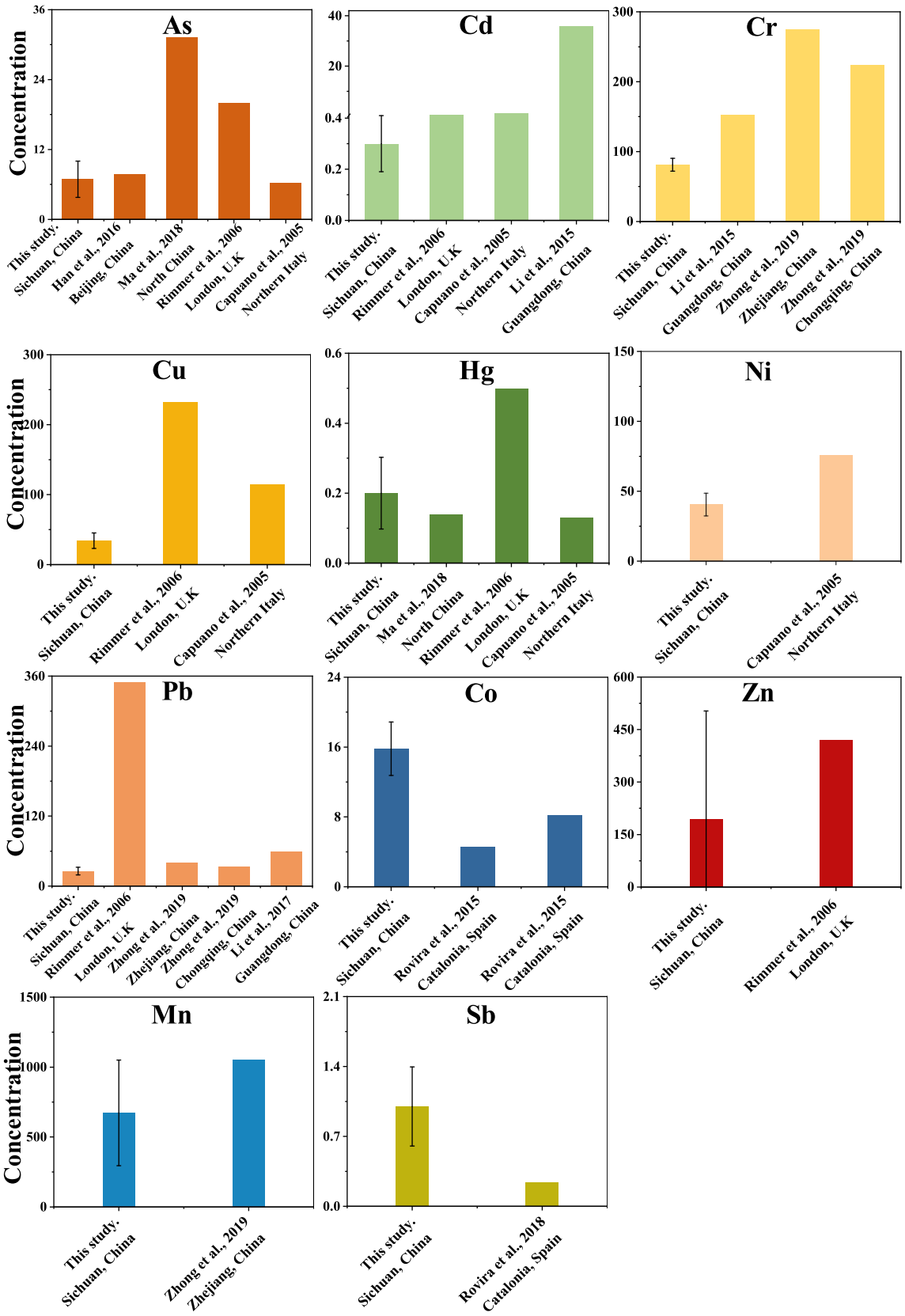


Fig. 4. Comparison of previously identified heavy metal soil concentrations (mg/kg) and those in soil near the MSW incineration plants.

concentrations of As, Cd, Cr, Cu, Ni, Pb, Zn, and Mn, were lower than those found in previous research. These heavy metals were detected at levels lower than those found in Beijing (Han et al., 2016), Guangdong (Li et al., 2017), Zhejiang (Zhong et al., 2019), Chongqing (Zhong et al., 2019) and London (Rimmer et al., 2006). However, the concentrations of Hg, Co, and Sb were higher than those detected in Northern China (Zhong et al., 2019), Northern Italy (Capuano et al., 2005), and Catalonia in Spain (Rovira et al., 2014; Rovira et al., 2015; Rovira et al., 2018). Such spatial disparities in the distribution of heavy metals might be associated with differences in the industrial sources and the quantities emitted (Li et al., 2015).

PCA was conducted to further investigate the sources of the heavy metals (Fig. S1). Details of the PCA method are given in the Supporting information. Four principal components could be extracted with eigenvalues greater than 1: PC1, PC2, PC3 and PC4. PC1 was dominated by Fe, Ni, Mn, Cr, Co, and Cu, with a variance contribution rate of 30.70%. These metals, especially Cr and Ni, have high natural background concentrations owing to the weathering of parent materials and pedogenesis (Ma et al., 2018). The variance contribution rate of PC2 was 25.90% and included Hg, Pb, and Zn, which are related to anthropogenic emission sources; in particular, Pb and Zn are usually considered as representative pollutants discharged from waste incinerators (Ma et al., 2018). The PC3 variance contributed 14.94%, with a large positive load of Cd and Co. High concentrations of Co (exceeding background levels) can be caused by human activities, such as the use of pesticides and chemicals, which highlights the need for strict quality controls on commercially available agrochemicals (Nisa and Sujatha, 2020). PC4 mainly included As and Ni. The high As concentration in soil might be associated with anthropogenic waste and industrial activities (e.g., sewage sludge and industrial discharge) (Zhao et al., 2014). Discharge from industrial factories in the study area, such as glass plants, perfume factories, pharmaceutical factories, and wood conversion factories, could have contributed to the As soil contamination.

3.3. Correlation and clustering analysis

To understand the relationships between PCDD/Fs and heavy metals in the soil, heatmaps were generated based on the correlation between the 11 investigated heavy metals and the 17 2,3,7,8-chlorine substituted PCDD/F congeners (Fig. 5). The patterns of correlation between the PCDD/Fs and heavy metals could be divided into three groups based on the correlation coefficient (r). Strong positive correlations were observed among the PCDD/Fs homologs, e.g., 1,2,3,4,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDF, 1,2,3,6,7,8-HxCDF, and OCDF ($r > 0.91$, $p < 0.05$). High correlations were also observed between heavy metals and PCDD/Fs. For example, Hg and Pb showed high positive correlations ($r > 0.51$, $p < 0.05$) with nearly all PCDD/Fs homologs (excluding 1,2,3,6,7,8-HxCDD and OCDD). Among the heavy metals, two groups were found highly correlated, e.g., Hg and Pb ($r = 0.83$, $p < 0.05$), Co and Mn, ($r > 0.91$, $p < 0.05$). A high correlation ($r > 0.51$, $p < 0.05$) between heavy metals and PCDD/Fs in the surface soils indicates that they may have originated from a similar emission source. Using the Pb isotope analysis method, Li et al. (2019b) found that coal combustion, vehicle exhaust, and MSW incineration were the three possible anthropogenic sources of Pb in soils surrounding MSW incineration plants. In addition, coal is typically added as a comburent during the start of when waste incineration, and coal combustion is a major contributor to Hg emission (Chen et al., 2016; Ma et al., 2018). The pollutant concentrations in soil might have been affected by waste incineration and the subsequent accumulation in soil via atmospheric deposition.

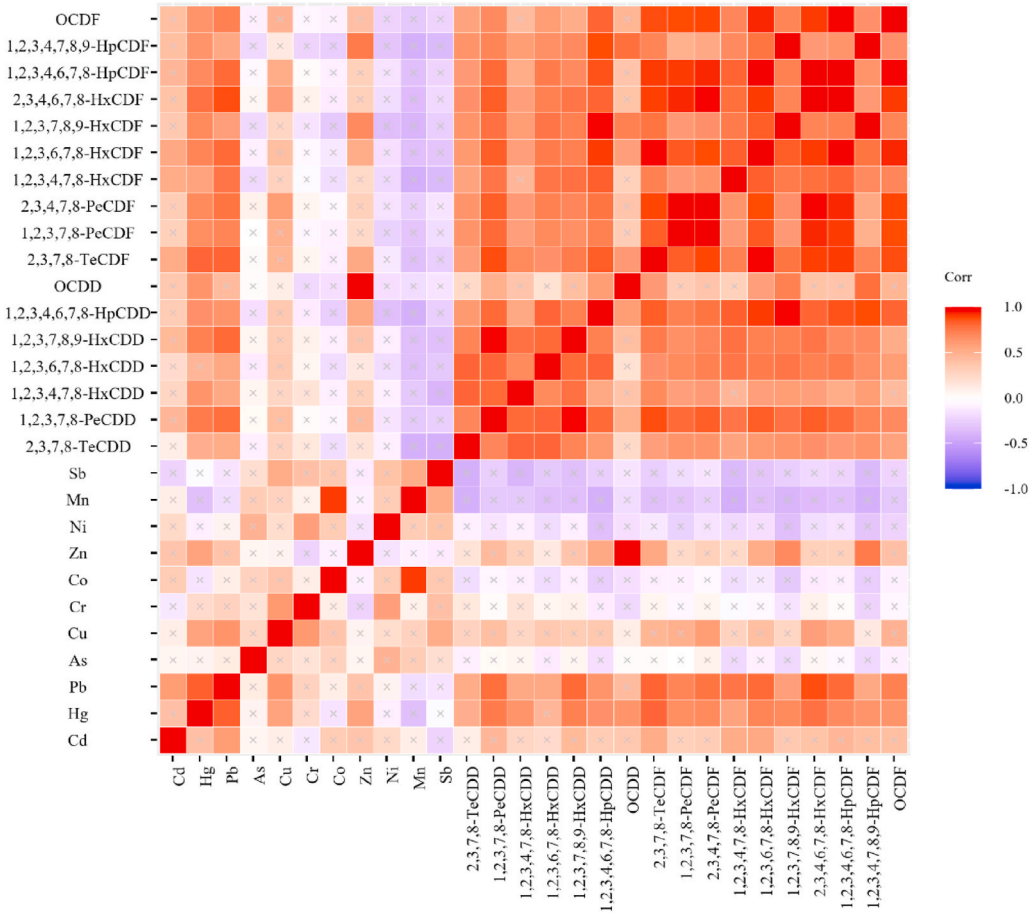


Fig. 5. Correlation analysis of the heavy metals and PCDD/Fs.

To further analyze the relationships between the heavy metals and PCDD/Fs, cluster analysis was carried out to categorize the species and identify analogous groups (Fig. S2). The PCDD/Fs and heavy metals were grouped into four clusters; compounds in the same cluster may have originated from similar emission sources. All of the species in Cluster 1 and Cluster 2 were heavy metals. Notably, Hg, Zn and Pb (identified as PC2 in the PCA results) were clustered in Cluster 3 and Cluster 4 with all of the PCDD/Fs homologs. As common wastes, batteries, paints, colored glass, waste tires, and inks used on plastics and paper contain high quantities of Cd, Zn and Pb (Zhang et al., 2008; Long et al., 2011; Yu et al., 2016). Therefore, the PCDD/Fs in the present study may have come from waste incineration emissions. Given the strong correlation between the heavy metals and the PCDD/Fs and their similar sources, as well as the large diversity of PCDD/Fs, high monitoring costs and complex detection technology required, these heavy metals could be used to characterize the distribution and concentrations of PCDD/Fs in soil. In the future, regularly monitoring the soil concentration for Hg, Cd, Zn and Pb could provide a reliable early warning for PCDD/Fs contamination arising from in MSW incineration.

3.4. Human health risk assessment

3.4.1. Health risk assessment of PCDD/Fs

The carcinogenic and non-carcinogenic risks were calculated based on the maximum PCDD/Fs inhalation exposure, dermal exposure, and ingestion level of people living in the vicinity of the MSW incineration plants. The *CR* values of the 17 detected PCDD/Fs for the three age groups (children, teenagers, and adults) associated with three exposure pathways are shown in Fig. 6(a–c). The total PCDD/Fs *CR* values for children, teenagers and adults were 5.68E-07, 1.45E-07 and 5.28E-07, respectively, which were far below the acceptable levels (1.00E-06 < *CR* < 1.00E-04) (US EPA, 2018). These results suggested there was no potential carcinogenic risk for these three age groups. Regarding the three exposure pathways, ingestion was the main route of soil exposure to PCDD/Fs in the study area, accounting for 78.51%, 64.80%, and 66.41% of the total exposure for children, teenagers, and adults, respectively.

The non-carcinogenic risks of PCDD/Fs exposure for the three age groups though the three exposure pathways are shown in Fig. 6(d–f). The total *HI* values of PCDD/Fs environmental exposure for children, teenagers, and adults were 4.83E-02, 4.74E-04, and 1.71E-02, respectively, which were far lower than the non-carcinogenic risk threshold (*HI* < 1), indicating no obvious non-carcinogenic risk. Compared with the *CR*, there were some differences in the distribution of the *HI* values in the three age groups. This was mainly because the average exposure time in the *CR* assessment for all the age groups was 70 × 365 days, while in the *NCR* assessment, the exposure times for children, teenagers, and adults were 11 × 365, 6 × 365, and 53 × 365 days, respectively. Regarding the three exposure pathways, ingestion was the main route of soil exposure to PCDD/Fs, accounting for 64.55%, 30.18% and 48.95% of the total exposure for children, teenagers, and adults, respectively.

A comparison with previous studies on the health risks of PCDD/Fs in soils near MSW incineration plants is shown in Table 2. The health risks (i.e., carcinogenic risk and non-carcinogenic risk) of PCDD/Fs for the three age groups in this study were similar to those found in Eastern China (Li et al., 2018), but they were significantly lower than the risks identified in Beijing (Hao et al., 2018) and Shanghai (Deng et al., 2020) and in the Spanish cities of Mataró (Rovira et al., 2015), Campdorà (Rovira et al., 2018), and Barcelona (Domingo et al., 2017). In general, the health risks posed by PCDD/Fs soil contaminations for the three age groups at all sample locations were below the acceptable levels, indicating no severe adverse human health effects.

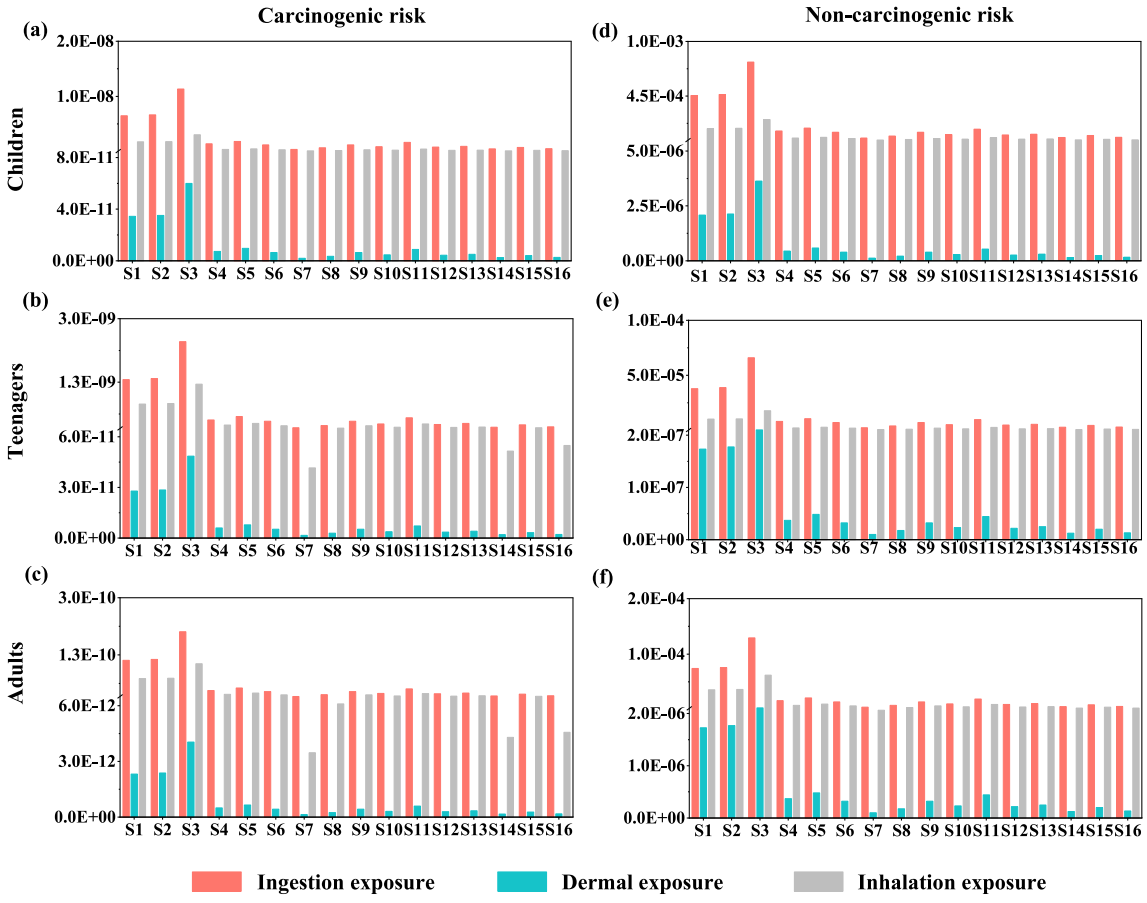


Fig. 6. Carcinogenic (**a-c**) and non-carcinogenic risks (**d-f**) from exposure to PCDD/Fs in soil near the MSW incineration plants.

Table 2
Comparison with previous studies regarding the health risks from PCDD/Fs in soil near the MSW incineration plants.

Country	Region	Receptor	Health risks		Reference
			HI	CR	
Spain	2011–2013, Mataró	Adults	8.75E-04	5.25E-07	Rovira et al. (2015)
	2015–2016, Campdorà	–	(1.00E-03, 1.00E-02)	(1.00E-07,1.00E-06)	Rovira et al. (2018)
	2014, Barcelona	–	2.00E-02	2.50E-06	Domingo et al. (2017)
	2017, Barcelona	–	1.00E-02	2.30E-06	
China	2008–2012, Beijing	Child	–	(6.80E-05, 3.20E-01)	Hao et al. (2018)
		Adult	–	(3.84E-05, 1.80E-01)	
	2017, Shanghai	Child	2.92E-02	1.75E-05	Deng et al. (2020)
		Adults	1.20E-02	7.18E-06	
	2018, East China	Child	(7.38E-03,2.16E-02)	(1.17E-07, 3.55E-07)	Li et al. (2018a)
		Teenagers			
		Adults			
	2020, Sichuan	Child	4.83E-03	5.68E-07	This study
		Teenagers	4.74E-04	1.45E-07	
		Adults	1.71E-02	5.28E-07	

3.4.2. Health risk assessment of heavy metals

The *CR* and *NCR* assessment results for children, men, and women exposed to heavy metals in soil are shown in Fig. 7. Sb, Mn, Cu, Hg and Zn were excluded from the *CR* assessment because slope factors were not available. The three exposure pathways for all heavy metals (excluding Ni) were at or below acceptable levels (1.00E-06 < *CR* < 1.00E-04). Only the dermal exposure pathway of Ni had carcinogenic risk values (4.87E-04, 1.22E-03, and 1.29E-03) for children, men, and women, respectively) that exceeded the upper acceptable level. Dermal exposure (excluding Cd and Pb because of a lack of carcinogenicity slope factors in the other pathways) was considered as the main exposure pathway, accounting for nearly 100% of the total exposure for children, men, and women.

For non-carcinogenic risk, the *HI* values for each heavy metal did not exceed the upper acceptable level (*HI* < 1) (Fig. 7(d–f)), indicating that the non-carcinogenic health risk of exposure to heavy metals in soil was acceptable. The average *HI* value was the highest for children, followed by that for women (1.45E-01) and men (1.38E-01), which was mainly owing to the high respiration rate of children, their frequent hand-mouth activity, and the high absorption rate of certain toxic elements by the gastrointestinal tract, which makes children more susceptible to environmental pollution (Li et al., 2014b; Ma et al., 2018). The dermal exposure pathway accounted for 74.21%, 83.29%, and 83.89% of the total exposure for children, men and women, respectively.

In summary, the carcinogenic risk and non-carcinogenic risk health risks posed by most heavy metals in soil for children, men and women

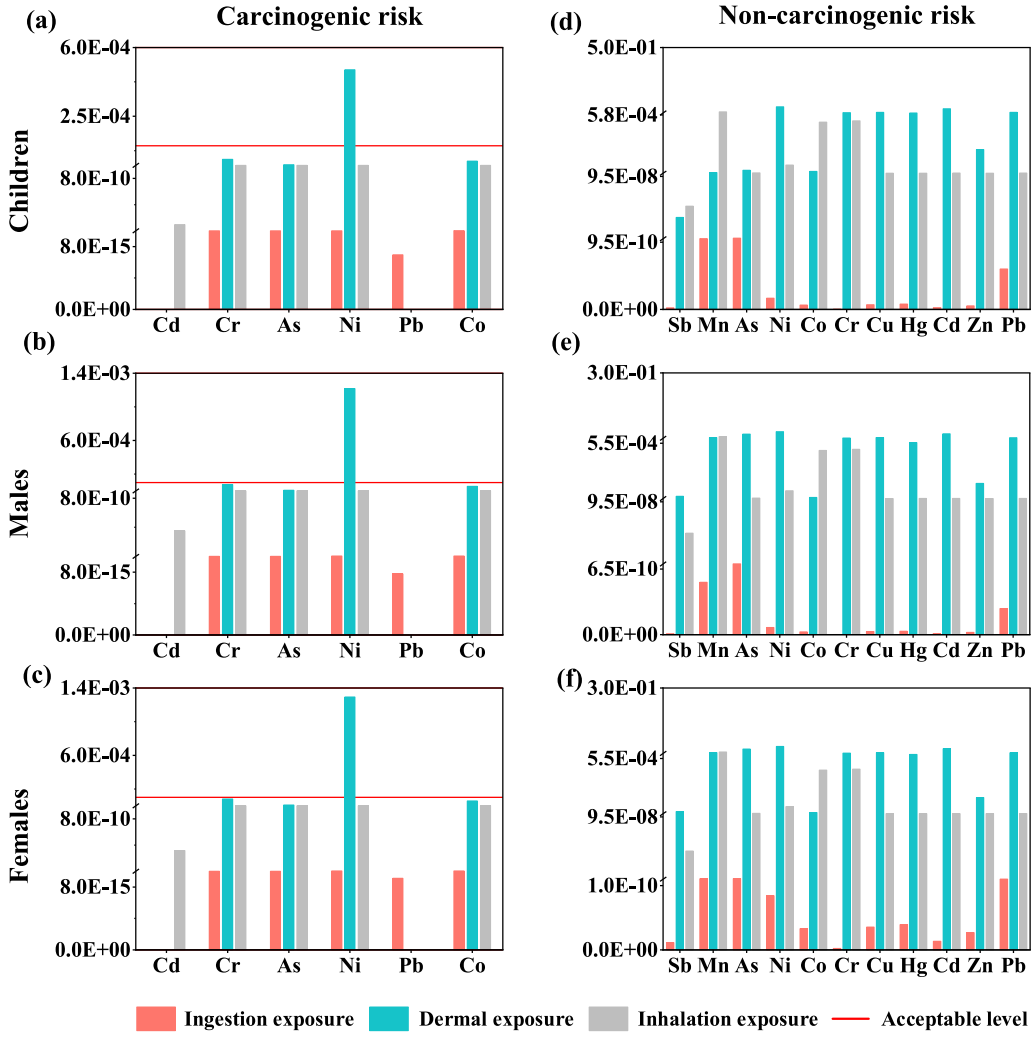


Fig. 7. Carcinogenic risk (**a-c**) and non-carcinogenic risk (**d-f**) derived from heavy metals exposure in soils around MSW incineration plants.

were all below acceptable levels, indicating no severe adverse human health effects. However, additional action is required to protect against the carcinogenic risk associated with Ni exposure from dermal contact with soil.

4. Conclusions

The waste incineration power generation industry is a potential emission source of heavy metals and PCDD/Fs, significantly impacting the surrounding soil. This study was the first comprehensive analysis of the concentration distribution of heavy metals and PCDD/Fs in soil near MSW incineration plants in Sichuan province. The correlations between heavy metals and PCDD/Fs were explored, and human health risks were assessed. The main findings were as follows: (1) The PCDD/Fs concentrations in all samples were low. The highest PCDD/Fs concentration (7.50 ng I-TEQ/kg) was found at sampling location S3, which was downwind of Plant A; however, it was significantly below the applicable risk screening (40 ng I-TEQ/kg) and risk intervention values (400 ng I-TEQ/kg). (2) The PCA results suggested that the principal components Hg, Pb and Zn (with a variance contribution rate of 25.90%), represented the characteristic metal pollutants emitted by the MSW incineration plants. (3) Regarding correlations among the pollutants, all PCDD/Fs homologs and heavy metals were clustered into one group, indicating that soil metal concentrations, especially of Hg, Pb, Zn and Cd, could be used as evidence for the accumulation of PCDD/Fs from MSW incineration plants. (4) The carcinogenic and non-carcinogenic health risks of PCDD/Fs and heavy metals (except for Ni) in the soils around the MSW incineration plants were within acceptable levels, indicating no severe, or extremely low, adverse human health effects. However, more attention is needed to prevent carcinogenic risk from Ni exposure through dermal contact with soil. The findings of this investigation will have an essential role in further population environmental epidemiology and environmental toxicology studies. We plan to continue investigating pollution characteristics and will conduct health risk evaluations in subsequent studies. Our future studies will consider the waste composition, operational conditions, and load capacities of waste incinerators, which have different roles in MSW incineration emissions. This information would be helpful when assessing soil pollution near MSW incineration plants and could enable us to obtain more accurate assessment results.

Author statement

Xin Bo: Conceptualization, Methodology, Writing-Original draft preparation. **Jing Guo:** Sample collection, Software, Writing- Reviewing and Editing. **Ruxing Wan**: Data curation, Visualization, Writing-Reviewing and Editing. **Yuling Jia:** Visualization, Investigation. **Zhaoxu Yang**: Sample collection, Validation. **Yong Lu**: Sample testing, Sample quality control. **Min Wei:** Supervision, Writing- Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.118816.

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