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PCDD/Fs and heavy metals in the vicinity of landfill used for MSWI fly ash disposal: Pollutant distribution and environmental impact assessment☆

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A B S T R A C T

This study focused on the syngenetic control of polychlorinated-ρ-dibenzodioxins and dibenzofurans (PCDD/Fs) and heavy metals by field stabilization/solidification (S/S) treatment for municipal solid waste incineration fly ash (MSWIFA) and multi-step leachate treatment. Modified European Community Bureau of Reference (BCR) speciation analysis and risk assessment code (RAC) revealed the medium environment risk of Cd and Mn, indicating the necessity of S/S treatment for MSWIFA. S/S treatment significantly declined the mass/toxic concentrations of PCDD/Fs (i.e., from 7.21 to 4.25 μg/kg; from 0.32 to 0.20 μg I-TEQ/kg) and heavy metals in MSWIFA due to chemical fixation and dilution effect. The S/S mechanism of sodium dimethyldithiocarbamate (SDD) and cement was decreasing heavy metals in the mild acid-soluble fraction to reduce their mobility and bioavailability. Oxidation treatment of leachate reduced the PCDD/F concentration from 49.10 to 28.71 pg/L (i.e., from 1.60 to 0.98 pg I-TEQ/L) by suspension absorption or NaClO oxidation decomposition, whereas a so-called "memory effect" phenomena in the subsequent procedures (adsorption, press filtration, flocculating settling, slurry separation, and carbon filtration) increased it back to 38.60 pg/L (1.66 pg I-TEQ/L). Moreover, the multi-step leachate treatment also effectively reduced the concentrations of heavy metals to 1–4 orders of magnitude lower than the national emission standards. Furthermore, the PCDD/Fs and heavy metals in other multiple media (soil, landfill leachate, groundwater, and river water) and their spatial distribution characteristics site were also investigated. No evidence showed any influence of the landfill on the surrounding liquid media. The slightly higher concentration of PCDD/Fs in the soil samples was ascribed to other waste management processes (transportation and unloading) or other local source (hazardous incineration plant). Therefore, proper management of landfills and leachate has a negligible effect on the surrounding environment.

1. Introduction

Incineration has become the major treatment method for municipal solid waste (MSW) in China, which disposes of over 62.3% of MSW (146.08 million tons) in 2020 (NBSC, 2021). The MSW incineration (MSWI) process will produce a large amount of fly ash, which generally accounts for approximately 5%–15% of the incineration capacity (Ma et al., 2021). The MSWI fly ash (MSWIFA) was classified as hazardous materials in many countries due to the multi-pollutants of heavy metals, soluble salts, and polychlorinated-p-dioxins and furans (PCDD/Fs), etc. (Chen et al., 2019; Peng et al., 2020).

The high concentration of potentially toxic heavy metals (Cd, Zn, Pb, Cr, Ni, etc.) in MSWIFA can be easily leached into the surrounding soil and groundwater if not treated properly (Pan et al., 2013; Wang et al., 2022a, b), causing significant harm to environment and human health (Bayuseno and Schmahl, 2011). For the raw MSWIFA, the leaching concentrations of Pb and Cd were reported frequently to exceed emission standards worldwide, and the high leaching concentrations of Zn and Cr were also observed (Vavva et al., 2017; Tong et al., 2019; Xu et al., 2019; Fan et al., 2021; Tian et al., 2021). Pan et al. (2013) reported the leached Cd in 67% of MSWIFA samples from 15 typical MSWI plants exceeded the Chinese regulated limitation, moreover, the risk

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assessment code (RAC) evaluated a high risk of Cd and Pb to environment. Hung et al. (2009) reported Pb and Cr leaching from MSWIFA landfill sites in Taiwan caused serious groundwater pollution. To avoid polluting the environment, landfill after stabilization/solidification (S/S) and resource utilization were the preferred treatment methods (Neramitkornburi et al., 2015; Wang et al., 2019; Marieta et al., 2021; Wang et al., 2021; Wang et al., 2022), wherein the landfill after S/S treatment was widely used (Liu et al., 2019; Luo et al., 2019). Cement S/S was a mature technology, which could transform MSWIFA into bulk or granular products, and the S/S products could be easily landfilled (Li et al., 2019; Chen et al., 2021). However, Liu et al. (2022) evaluated and predicted that the cumulative leaching amount of Pb in 718 days was higher than the national limit in GB16889-2008 (MEEPRC, 2008a), while the leaching amount of Cu, Cr, As and Ni did not exceed that even in 50 years. To prevent the leaching of heavy metals into environment, some inorganic or organic chelating reagents were applied in S/S process, such as phosphate, silicates, sulfides, ethylene diaminetetracetic acid (EDTA), tetrathio bicarbamic acid (TBA), sixthio guanidine acid (SGA), etc. (Wang et al., 2015; Ma et al., 2019). These additives could convert heavy metals to physically and chemically stable forms. In China, combining cement with reagents was a common pre-treatment method for MSWIFA before landfill (Li et al., 2018a, b; Zhang et al., 2020). However, few studies evaluated its environmental impact in the vicinity of landfill sites, and it is urgent to fill relevant understanding gaps.

The PCDD/F concentration in MSWIFA was another key issue. The PCDD/Fs with very low water solubility, which belongs to hydrophobic organic pollutants (HOP), were considered very difficult to leachate from the contamination sources (Choi and Lee, 2006). However, Wang et al. (2006) found that the PCDD/F concentrations in the groundwater and the treated leachate in a landfill site for solidified MSWIFA were all one order of magnitude higher than those in the background samples, indicating landfill sites without proper management could influence the surrounding environment. In addition, the PCDD/F concentration in the surface soils of the landfill site was 460 times higher than that of urban soils and the highest value was 2.8 times higher than the Taiwan soil regulation (1 μg I-TEQ/kg), although the leaching concentrations of PCDD/Fs were extremely lower than the local regulation (Wang et al., 2006). Nevertheless, the PCDD/F concentrations in leachate and surrounding media of landfill sites were less concerned recently, and its environmental risk could not be ignored.

In addition to the S/S treatment of MSWIFA, the following disposal of the leachate produced from landfill site was also essential. The untreated leachate could pollute the groundwater, soil, and other environmental media due to the high concentration of organic pollutants, heavy metals, inorganic salts, etc. Previous studies developed many treatment methods for landfill leachate, and some so-called conventional technologies have been applied to landfill sites, including oxidation, coagulation, flocculating, adsorption, and filtration (Carvajal-Flórez and Santiago-Alonso, 2019; Gautam et al., 2019; Babaei et al., 2021; Lin et al., 2022). These treatment processes were mainly designed for the emission control of heavy metals, dissolved inorganic matter, and dissolved organic matter, however, less concern was focused on the PCDD/Fs.

This study focused on two key pollutants (PCDD/Fs and heavy metals) in the vicinity of landfill used for MSWIFA disposal, especially the pollutant distribution characteristics and the environmental impact assessment. The major investigation media around landfill sites included MSWIFA, soil, groundwater, leachate, and river water. In addition, the S/S treatment technology for MSWIFA and the multi-step treatment for leachate were investigated for their synergistic implication in controlling heavy metals and PCDD/Fs. Based on these, this study further evaluated the environmental behavior, bioavailability, and risk assessment of heavy metals and PCDD/Fs.

2. Materials and methods

2.1. Landfill site

The landfill site located in Hangzhou, China, surrounded by hills (approximately 200 m) on three sides (north, west, and south) (Fig. S1). The wind generally blows into the landfill from the east. The shortest distance between the landfill site and the nearest river is approximately 1.8 km, separated by a mountain (approximately 360 m). 200 m southeast of the landfill site is an environmental service company, which has two rotary kilns incineration systems (1.0 t/h and 2.4 t/h, respectively) for hazardous waste treatment. This landfill site has a storage capacity of 0.123 million m^3 with an operation period of over 7 years. It is an anaerobic landfill site, and organic matter in landfill wastes was controlled less than 5%. The liner structure of the landfill was composed of two infiltration proof layers and two layers of silty clay according to the technical code for landfill sites in China (MHURDPRC, 2007). The disposal waste was mainly the fly ash produced from local circulating fluidized bed (CFB) MSWI and hazardous waste incineration (HWI) plants. The proportions of landfill wastes were approximately distributed as MSWIFA (65%), HW fly ash (6%), bottom ash of HW (17%), sewage sludge (10%), and others (2%). Other wastes, such as garbage, combustible waste, incombustible waste, etc., were not allowed to be landfilled on this site. The generated leachate was treated by multiple purification steps, including oxidation, adsorption, press filtration, coagulation/flocculating, slurry separation, and carbon filtration. The oxidation step was conducted in a reactor with a stirring, and the oxidant was sodium hypochlorite (NaClO). The solid residue from filtration will be disposed of in-site, and the treated leachate was finally disposed of off-site.

2.2. Samples collection

In this study, the untreated MSWIFA was collected from the bag filter of a CFB MSWI system. The S/S method was adding organic chelate reagent (Sodium dimethyldithiocarbamate, $C_3H_6NNaS_2$, SDD, 1%) and cement (10%), then the S/S products were packed and landfilled in-site (details in Supplementary Material). All samples (MSWIFA, leachate, groundwater, river water, and soil) were collected in the same sampling batch followed the Chinese standards of HJ 77.1–2008, HJ 77.3–2008, and HJ 77.4–2008 (MEEPRC, 2008b,c,d). The sampling procedures were conducted by the technical team from the Dioxin Laboratory, Zhejiang University (China). The MSWIFA samples were collected with two replications before and after S/S procedures from the landfill pretreatment workshop. The leachate samples (1 L) were collected with two replications from 0.5 m depth in landfill leachate ponds (original, after oxidation, final), corresponding to sampling sites 1, 2, and 3 marked in Fig. S1. The groundwater samples (1 L) were collected with two replications from the upstream, downstream, and background monitoring wells (sampling sites 4, 5, and 6). The river water samples were obtained in sampling site 7, which was 2.4 km away from the landfill site. All liquid samples were stored in glass bottles, which were washed with HCl (10%) to avoid foreign ions before each sampling. The soil samples consisted of topsoil approximately 20 cm from the center point and topsoil of 20 cm each at four additional points 5–6 m from the center point in 4 main directions (N, E, S, W). The soil samples were obtained near the landfill sites (sampling sites 9 and 10), while the background soil was collected in an ecological park due to less human activity (sampling site 8). All samples were stored in airtight glass bottles under dark conditions, kept at a low temperature during transportation, and analyzed within 72 h. Detailed information on different samples were summarized in Table 1 and Fig. S1.

2.3. Samples analysis

The PCDD/Fs in different samples (MSWIFA, leachate, groundwater,

Table 1
Summary information of the liquid and solid samples from the landfill site.

Sample type	Sample code	Phase	Sample description	Sampling sites
MSWIFA	**FA-UT**	Solid	Untreated MSWIFA	/
	FA-T	Solid	S/S treated MSWIFA	/
Leachate	**L-UT**	Liquid	Untreated leachate	1
	L-PT	Liquid	Preliminarily treated leachate after oxidation	2
	L-T	Liquid	Treated leachate	3
Groundwater	**GW-U**	Liquid	Groundwater in upstream monitoring well	4
	GW-D	Liquid	Groundwater in downstream monitoring well	5
	GW-B	Liquid	Groundwater in background monitoring well	6
River water	**RW**	Liquid	River water	7
Soil	**S–B**	Solid	Background soil sample	8
	S-DW	Solid	The predominant wind direction was from the east, and this soil sample was collected on the downwind side and southwest edge of the landfill area	9
	S-NL	Solid	Soil sample near the leachate treatment area	10

river water, and soil) were pretreated and analyzed according to the Chinese standards of HJ 77.1–2008, HJ 77.3–2008, and HJ 77.4–2008 (MEEPRC, 2008b,c,d). All experimental solvents of pesticide residue analysis grade were purchased from Mallinckrodt Baker Inc., USA. The detailed procedures of PCDD/F analysis had been described in our previous works (Chen et al., 2014; Chen et al., 2017). The international toxic equivalent factor (I-TEF) scheme for PCDD/Fs was used to calculate the international toxic equivalency (I-TEQ) (Bhavsar et al., 2008). A high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS) (JMS-800D, JEOL, Japan) with a DB-5MS (60 m length × 0.25 mm internal diameter × 0.25 μm film) column was used to analyze the PCDD/Fs in the different samples. The quality control and quality assurance were assured by using $^{13}C_{12}$-labeled internal standards in each sample during the strict pretreatment procedures (extraction, rotary evaporator, cleanup by multilayer silica gel column and alumina column, nitrogen-blowing), which guaranteed the unequivocal identification of the target compounds and the recoveries check of different congeners. In addition, regular spectrometer testing (sensitivity and reproducibility) by analysis of analytical standards and laboratory blanks was periodically carried out. A parallel test was conducted every 10 samples, and the error was within 20%. Furthermore, the laboratory owned the China Inspection Body and Laboratory Mandatory Approval (CMA) and regularly participated in international laboratory comparisons for the analysis of PCDD/Fs to ensure the quality of data obtained. In this study, the recoveries of PCDD/F standards ranged from 33.4% to 124.7%, fulfilling the standard requirements (MEEPRC, 2008b,c,d).

The total concentration (C_t) of targeted heavy metals in the untreated and treated MSWIFA was determined by microwave digestion method according to the Chinese standard of HJ 781-2016 (MEEPRC, 2016). The MSWIFA sample was analyzed in triplicate, and the results reported in this study were the average values with standard deviation. A modified five-step sequential European Community Bureau of Reference (BCR) extraction procedure (Table S1) was used to examine the speciation of heavy metals in the MSWIFA samples. According to the Chinese standard of GB/T 25282-2010 (MEEPRC, 2010), the concentrations of heavy metals in MSWIFA (Zn, Pb, Cu, Cr, Ni, Cd, Ba, Be, As, Co, and Mo) were detected by using inductively coupled plasma atomic emission spectrometry (ICP-AES, Thermo iCAP 6300, USA), and the concentrations of heavy metals (Hg, Sb, and Se) were detected by Atomic fluorescence spectrometry (AFS, XGY1011A, China). The concentrations of heavy metals in liquid samples (Zn, Pb, Cu, Cr, Ni, Cd, Ba, As, Se, Be, and Hg) were detected by the inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500a, USA) according to the Chinese standard of HJ 700-2014 (MEEPRC, 2014). The 1,5-Diphenylcarbohydrazide spectrophotometric method was used for the determination of Cr(Ⅵ) in liquid samples (MEEPRC, 1987).

2.4. Statistical analysis

To better evaluate the distribution of PCDD/F congeners, the chlorination degree of PCDD/Fs (d_{Cl}, the average number of chlorine substituents) was calculated as follows:

$$d_{Cl} = \frac{\sum C_j \times n_j}{C} \tag{1}$$

where $j = 4, 5, 6, 7, 8$, C_j represents the concentration of PCDDs, PCDFs, or PCDD/Fs, n_j represents the number of substituted chlorines in PCDDs, PCDFs, or PCDD/Fs; and C represents the total concentration of PCDDs, PCDFs, or PCDD/Fs.

In the BCR sequential extraction process, to effectively recognize the environmental behavior and bioavailability of heavy metals and potentially toxic metals, the five fractions in the FA sample were usually analyzed, namely, the mild acid-soluble fraction (F1), the reducible fraction (F2), the oxidizable fraction (F3), the residual fraction (F4), and water-soluble (F5).

Serving as an internal check for the precision of these methods, the recovery rate of targeted metals (REC, %, calculated by Eq. (2)) was determined by comparing the sum of the four fractions (F1, F2, F3, and F4) with the total concentration of heavy metals in MSWIFA.

$$REC = \frac{F1 + F2 + F3 + F4}{C_t} \times 100 \tag{2}$$

The risk assessment code (RAC) of the targeted metals (%) was defined as the fraction of acid-soluble elements in the total species and was calculated by Eq. (3),

$$RAC = \frac{F1}{F1 + F2 + F3 + F4} \times 100 \tag{3}$$

RAC was usually graded into five levels to indicate the environment risk, including no risk (<1%), low risk (1–10%), medium risk (10%–30%), high risk (30–50%) and very high risk (>50%) (Pan et al., 2013).

The S/S efficiency of heavy metals (%) was introduced, which was defined as the reduction effect of heavy metals in mild acid-soluble fraction (F1), and was calculated by Eq. (4),

$$S/S\ efficiency = \frac{C_{F1-UT} - C_{F1-T}}{C_{F1-UT}} \times 100 \tag{4}$$

where the C_{F1-UT} represents the F1 concentrations of heavy metals in untreated MSWIFA, and the C_{F1-T} represents those in treated MSWIFA.

3. Results and discussion

3.1. The distribution of PCDD/Fs

Table 2 summarized the PCDD/F characteristics in different media in the vicinity of the landfill site. The original PCDD/F concentrations in MSWIFA was 7.21 μg/kg (0.32 μg I-TEQ/kg), which had met the national landfill standard of MSWIFA (3.0 μg I-TEQ/kg in GB 16889-2008) (MEEPRC, 2008a). The S/S treatment further decreased the concentration to 4.25 μg/kg (0.20 μg I-TEQ/kg). The PCDD/Fs were hardly oxidized and destructed in the S/S process. According to the additional amount of SDD (1%) and cement (10%), the dilution effect of the S/S process was the major reason. Thus, the chlorination degree and congener distribution of PCDD/Fs only showed a slight change after S/S treatment (Table 2, Fig. 1a and b). The high-chlorinated congeners of PCDD/Fs ($d_{Cl} \geq 6$) were still the dominant compounds, accounting for

Table 2
PCDD/F concentrations of solid and liquid samples (μg/kg or μg I-TEQ/kg for MSWIFA and soil samples; pg/L or pg I-TEQ/L for liquid samples).

Samples	PCDDs	PCDFs	PCDD/Fs	PCDD/Fs (I-TEQ)	PCDFs/(PCDD/Fs)	$d_{Cl-PCDDs}$	$d_{Cl-PCDFs}$	$d_{Cl-PCDD/Fs}$
FA-UT	**2.80 ± 0.27**	**4.41 ± 0.19**	**7.21 ± 0.23**	**0.32 ± 0.012**	0.61	7.55	6.75	7.06
FA-T	**1.29 ± 0.20**	**2.96 ± 0.33**	**4.25 ± 0.28**	**0.20 ± 0.011**	0.70	7.47	6.82	7.02
L-UT	**32.13 ± 1.09**	**16.97 ± 1.24**	**49.10 ± 1.12**	**1.60 ± 0.093**	0.35	7.54	6.58	7.21
L-PT	**13.93 ± 0.86**	**14.77 ± 0.98**	**28.71 ± 0.96**	**0.98 ± 0.096**	0.51	7.65	6.74	7.18
L-T	**19.84 ± 1.24**	**18.76 ± 1.10**	**38.60 ± 1.19**	**1.66 ± 0.13**	0.49	7.43	6.72	7.09
GW-U	**17.56 ± 1.31**	**4.29 ± 0.19**	**21.84 ± 1.16**	**0.28 ± 0.023**	0.20	7.90	6.80	7.68
GW-D	**2.84 ± 0.21**	**3.34 ± 0.33**	**6.19 ± 0.29**	**0.23 ± 0.011**	0.54	7.57	6.80	7.15
GW-B	**2.92 ± 0.19**	**3.38 ± 0.12**	**6.30 ± 0.14**	**0.18 ± 0.0091**	0.54	7.84	6.85	7.31
RW	**13.11 ± 1.05**	**3.63 ± 0.23**	**16.73 ± 1.01**	**0.31 ± 0.015**	0.22	7.80	6.42	7.50
S–B	**0.12 ± 0.0031**	**0.013 ± 0.0051**	**0.14 ± 0.011**	$\mathbf{1.66 \times 10^{-3} \pm 1.32 \times 10^{-4}}$	0.10	7.91	6.61	7.78
S-DW	**7.85 ± 0.93**	**0.049 ± 0.0069**	**7.90 ± 0.53**	$\mathbf{1.63 \times 10^{-2} \pm 2.31 \times 10^{-4}}$	0.01	7.96	6.84	7.96
S-NL	**3.30 ± 0.18**	**0.029 ± 0.0026**	**3.33 ± 0.51**	$\mathbf{6.64 \times 10^{-3} \pm 9.54 \times 10^{-5}}$	0.01	7.98	6.76	7.97

92.25–92.55% (Fig. 1a). While the low-chlorinated congeners ($d_{Cl} \leq 5$) dominated the toxic concentration of PCDD/Fs, i.e., the 2,3,4,7,8-PeCDF individually contributed most (35% and 37%, respectively for untreated and treated MSWIFA) (Fig. 1b), and the 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD contributed 11% together. During the S/S process, the ratio of PCDFs increased from 0.61 to 0.70 due to the variation. Generally, the S/S process reduced the PCDD/F concentrations in MSWIFA, and it slightly influenced the distribution of PCDD/F congeners.

In this landfill site, the PCDD/F concentration in untreated leachate was 49.10 pg/L (1.60 pg I-TEQ/L), which was dominated by PCDDs (i.e., 32.13 pg/L; 65.4%) and high-chlorinated congeners (i.e., $d_{Cl-PCDD/Fs}$ = 7.54; 93.3%) (Table 2, Fig. 1c). After the oxidation treatment, the PCDD/F concentration decreased to 28.71 pg/L (0.98 pg I-TEQ/L), corresponding to the significant reduction of PCDDs (18.20 pg/L) (Table 2). It could be ascribed either to the absorption and depression of the suspension or the oxidation and decomposition by NaClO. On one hand, the PCDDs were easier to be adsorbed than PCDFs due to the lower vapor pressures (Mader and Pankow, 2003; Li et al., 2004); on the other hand, the PCDD were also easier to be oxidized and decomposed than PCDFs, wherein PCDDs had two oxygen atoms in their central ring instead of one in PCDF and the bond energy of C–O was also lower than C–H (Debecker et al., 2007). After the subsequent treatment procedures (adsorption, press filtration, flocculating settling, slurry separation, and carbon filtration), the PCDD/F concentration increased back to 38.60 pg/L (1.66 pg I-TEQ/L), in which PCDDs and PCDFs contributed 5.91 and 3.99 pg/L, respectively. Generally, leachate treatment only slightly influenced the proportions of high-chlorinated congeners (from 93.3% to 91.08% and 92.54%, respectively for L-UT, L-PT, and L-T) on mass concentrations, but the d_{Cl} and the dominative OCDD continuously decreased from 42% to 36% and 32%, respectively (Fig. 1c). The oxidation treatment showed less influence on the congener distribution of toxic concentrations, while the subsequent procedures increased the proportion of low-chlorinated congeners, especially the 2,3,7,8-TCDD (from 0 to 22%) (Fig. 1d). This study regarded this phenomenon as a kind of "memory effect", which could be attributed to the accumulation in the procedure of "adsorption, press filtration, flocculating settling, slurry separation, and carbon filtration". The accumulated PCDD/Fs were released into leachate when the accumulated concentration of PCDD/Fs was high enough. The accumulated PCDD/Fs could attribute to less cleaning of the leachate treatment pipeline (Fig. S1), which provided large surface for PCDD/F storage. Our group reported similar phenomena in the wet scrubbing systems equipped in MSWI plants (Ma et al., 2019; Lin et al., 2020a,b; Ma et al., 2020). Nevertheless, these concentrations of PCDD/Fs in leachates were all below the regulation level of PCDD/F concentrations in leachates by pure water (10 pg I-TEQ/L) (Yasuhara and Katami, 2007). The leachate treatment techniques can reduce the original emission of PCDD/Fs.

For the groundwater, the PCDD/F concentration in upstream groundwater was 21.84 pg/L (0.28 pg I-TEQ/L), which was dominated by PCDDs (i.e., 17.56 pg/L; 80.4%) and high-chlorinated congeners (i.e., $d_{Cl-PCDD/Fs}$ = 7.68; 97.3%) (Table 2, Fig. 1e). The PCDD/F concentration decreased to 6.19 pg/L in downstream groundwater, and the PCDD concentration decreased to 2.84 pg/L. Although the PCDD/Fs were still dominated by high-chlorinated congeners ($d_{Cl-PCDD/Fs}$ = 7.15; 92.7%), the contributions of different congeners changed much, leading to less difference between PCDD and PCDF (Fig. 1e). The toxic concentration in GW-D (0.23 pg I-TEQ/L) was only slightly lower than that in GW-U due to the high proportion of OCDD (74%) and its low I-TEF value, and the congeners distribution based on toxic concentrations showed less change (Fig. 1f). Note that the toxic concentration in leachate was approximately 5 times higher than that in groundwater. All of which not only indicated the landfill site was not releasing PCDD/Fs into groundwater and was not the input source of PCDD/Fs but also suggested proper control and management of landfills and leachate would not influence the surrounding environment. The decreased PCDD/Fs might be ascribed to the transformation to surrounding aquitard or the dilution caused by the influx of other groundwater. The concentration of PCDD/Fs in GW-B was 6.30 pg/L (0.18 pg I-TEQ/L), which was also dominated by high-chlorinated PCDD/Fs (i.e., $d_{Cl-PCDD/Fs}$ = 7.31; 93.4%) and showed similar distribution of PCDD/F congeners in GW-D (Table 2 and Fig. 1e). As for the river water, the PCDD/F concentration was 16.73 pg/L (0.31 pg I-TEQ/L), which was dominated by PCDDs (i.e., 13.11 pg/L; 78.4%) and high-chlorinated congeners (i.e., $d_{Cl-PCDD/Fs}$ = 7.50; 94.0%). The distribution properties of PCDD/F congeners in GW-U and RW were similar (Table 2, Fig. 1e and f), however, originated from different sources. The major source of PCDD/Fs in RW could attribute to pesticide use in upstream agricultural land (Fig. S1). The PCDD/F source of GW-U could originate from the surrounding thermal industries, including casting and forging enterprises and plastic manufacturing (2–3 km away) (Fig. S1). In addition, the similar distribution properties of PCDD/F congeners between GW-D and GW-B may attribute to the nearby HWI plant. Similar results and these potential sources of PCDD/Fs were reported in other regions (Thuan et al., 2011; Grant et al., 2015; Nguyen et al., 2017).

For the soil, the PCDD/F concentration in background soil was 0.14 μg/kg (1.66×10^{-3} μg I-TEQ/kg), which was dominated by PCDDs (i.e., 0.12 ng/kg; 90.4%), especially OCDD (85%) (Table 2, Fig. 1g). However, OCDD only contributed 7% to the toxic concentration, while low-chlorinated congeners contributed 56.4% (Fig. 1h). In the southwest landfill area and downwind side, the PCDD/F concentration in S-DW was nearly 60 times higher than that of S–B (i.e., 7.90 μg/kg; 1.63×10^{-2} μg I-TEQ/kg). In the soil near the leachate treatment area, the PCDD/F concentration was 3.33 μg/kg (6.64×10^{-3} μg I-TEQ/kg). The higher PCDD/F concentration of S-DW could attribute to the specific position, which not only existed on the edge of the landfill area but was also located in the predominant wind of the landfill site. In addition, for

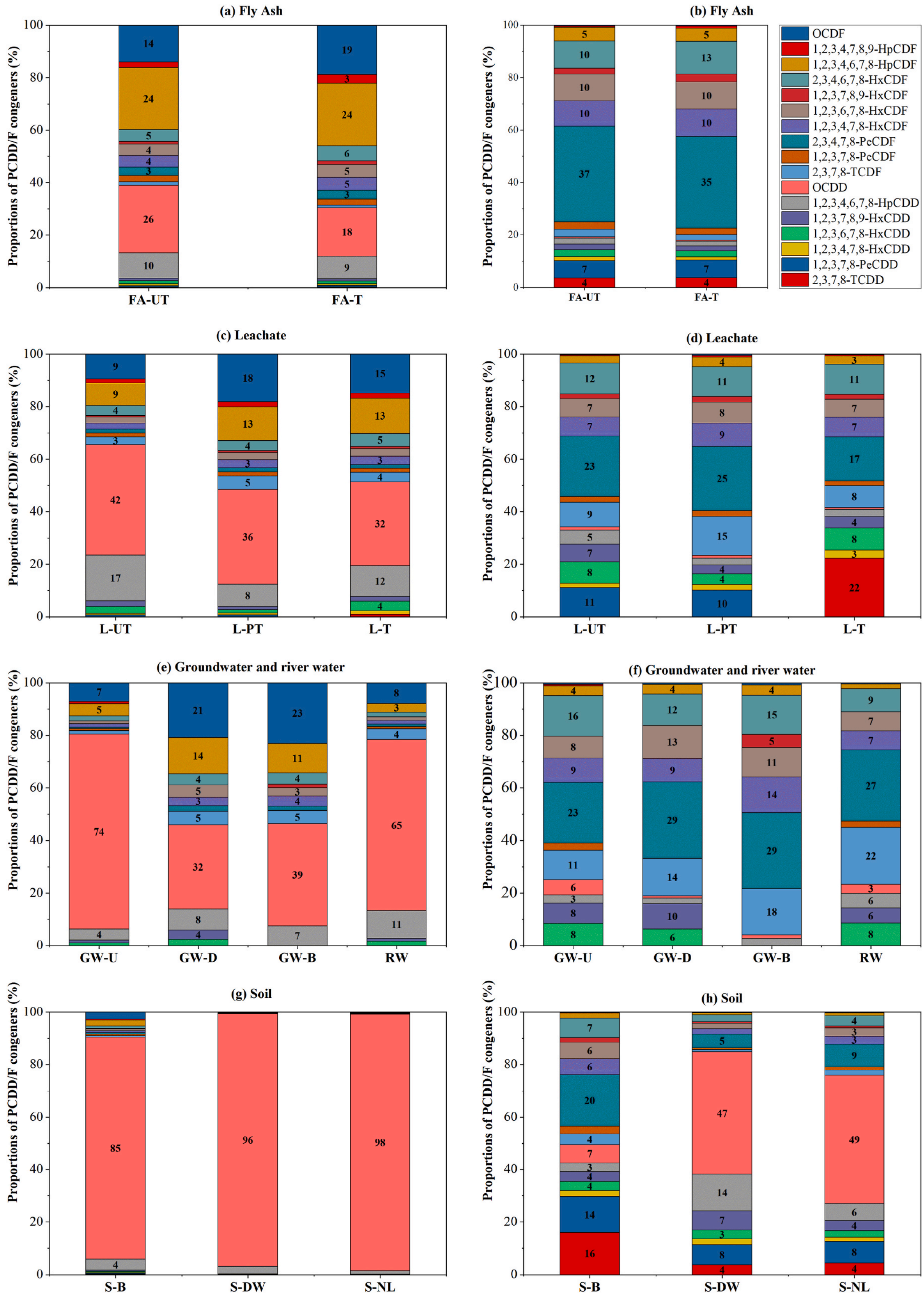

Fig. 1. Proportions of PCDD/F congeners based on total concentrations (a, c, e, and g) and toxic concentrations (b, d, f, and h).

S-DW and S-NL, OCDD was the dominant congener (i.e., 96% and 98%) with a slight discrepancy, which also led to a high chlorination degree (i.e., $d_{Cl-PCDD/Fs} = 7.94 - 7.97$) (Table 2) and the highest contribution to the toxic concentration (34% and 49%) (Fig. 1h). However, OCDD (85%) in S–B only contributed 7% to the toxic concentration, indicating the extra OCDD in S-DW and S-NL came from nearby. Previous studies also confirmed similar phenomena and regarded OCDD as the characteristic congener of PCDD/Fs in soil samples (Wang et al., 2006; Di Guardo et al., 2017). Furthermore, Wang et al. (2006) reviewed and concluded that the PCDD/Fs (especially the high-chlorinated congeners) possess such strong hydrophobicity and exhibit little downward mobility once deposited in or on soil, indicating less potential for soil to be a PCDD/F source of the nearby water environment. The high concentrations of PCDD/Fs in the S-DW and S-NL could be influenced by waste management processes (transportation and unloading) (Gworek et al., 2013) and other local sources, i.e., the hazardous incineration plant (Li et al., 2017, 2018; Lin et al., 2020a,b).

3.2. Heavy metals

3.2.1. Total concentration of heavy metals

As shown in Table S2, the concentrations of 14 heavy metals in untreated and S/S treated MSWIFA were detected. And the recovery rates of targeted metals (REC) were within the range of 100%–102.18% and 92.87%–100%, respectively. The untreated MSWIFA had a relatively high concentrations of heavy metals, and their concentrations (mg/kg) followed the order of Ba (9480.31) > Zn (5343.23) > Pb (3749.47) > Cu (1740.65) > Cr (1066.72) > Mn (823.92) > Ni (142.53) > Mo (126.31) > Cd (62.78) > Sb (57.66) > As (44.11) > Co (32.42) > Hg (27.05) > Be (1.07). The MSWIFA was mainly dominated by Ba, Zn, Pb, and Cu. The concentration of Ba was relatively high compared with some other samples in China, which could attribute to the CFB source of MSWIFA and the sample characteristics (Qiu et al., 2017; Fan et al., 2022). After the S/S treatment, the concentrations of heavy metals in MSWIFA significantly decreased (Table S2), mainly attributed to the chemical fixation and dilution effect.

3.2.2. BCR speciation analysis

Five fractions, i.e., the mild acid-soluble fraction (F1), the reducible fraction (F2), the oxidizable fraction (F3), the residual fraction (F4), and the water-soluble (F5), were analyzed (Table S2) and their proportions were shown in Fig. 2. The concentrations of all heavy metals in each fraction were decreased after the S/S treatment process (Table S2). The sodium dimethyldithiocarbamate ($C_3H_6NNaS_2$, SDD) can capture heavy metals with a single dithiocarboxy chelating group and form stable heavy metal chelate compounds (Wang et al., 2015; Zhang et al., 2020;

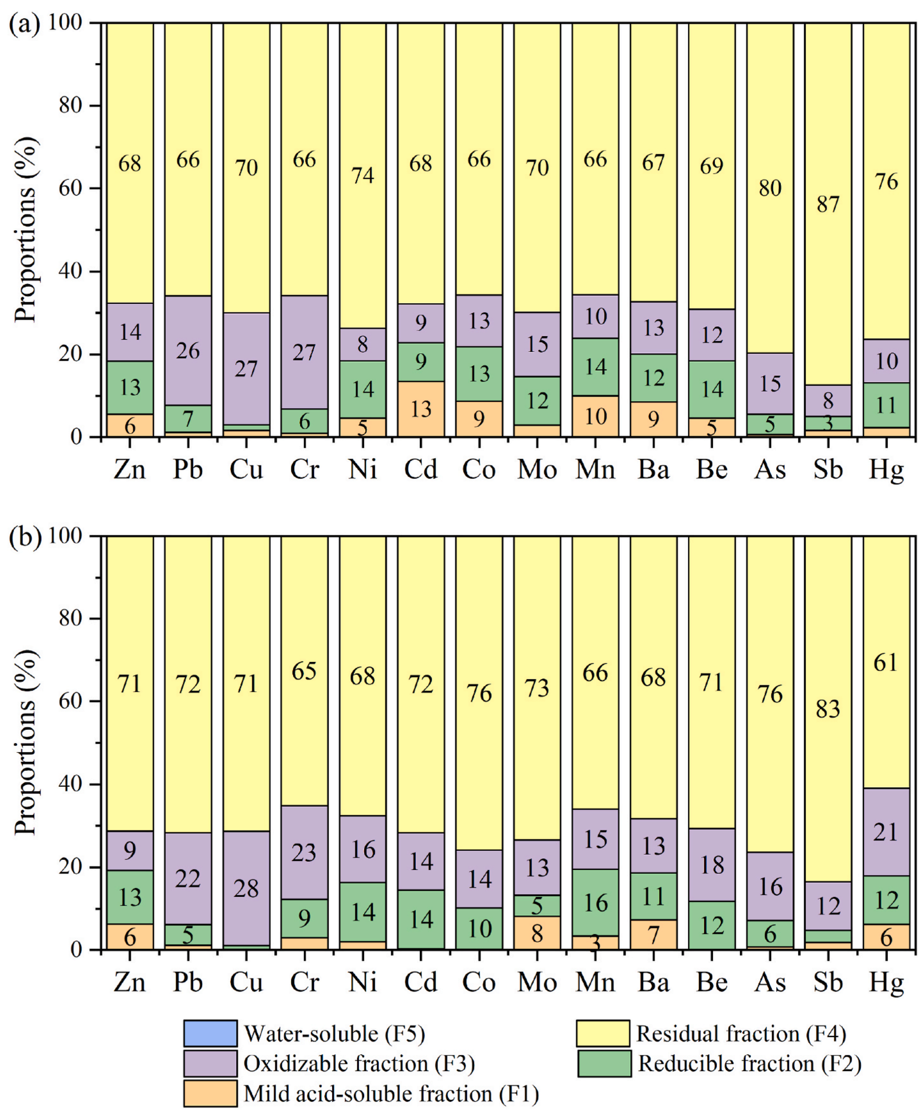

Fig. 2. The proportion of heavy metal species in the MSWIFA samples determined by BCR, (a) untreated and (b) S/S treated.

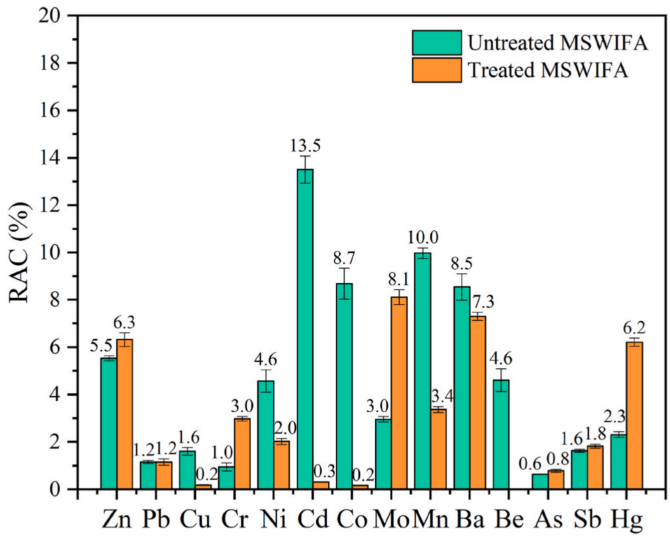

Fig. 3. Risk assessment code (RAC) of heavy metals in MSWIFA.

Wang et al., 2022). The addition of cement will not only further provide large amounts of alkalinity to stabilize heavy metals, but also form a harder cement solidified body to reduce their leaching risk by controlling their diffusion, surface wash-off, and dissolution (Wang et al., 2015; Liu et al., 2022; Wang et al., 2022). Ba, the most abundant heavy metal in untreated MSWIFA, in each fraction was dramatically reduced, while Zn became the most abundant heavy metal in S/S treated MSWIFA (Table S2). These could attribute to the field S/S techniques, which was not mixed well enough and caused the sampling variation. Hence, a longer mixing duration and premixture of MSWIFA and cement were suggested to improve the uniformity of the S/S mixture. The S/S treatment also impaired the leaching amounts of heavy metals in water-soluble fraction, resulting in a negligible contribution (Fig. 2).

Fig. 2 illustrated heavy metals were mainly distributed in the residual fraction in both untreated MSWIFA (i.e., 66%–87%) and in treated MSWIFA (i.e., 61%–83%). Residual fractions were mainly retained in minerals (e.g., quartz), which were not easily released under natural conditions (Wang et al., 2015). S/S treatment further increased the proportions of most heavy metals, i.e., Zn, Pb, Cu, Cd, Co, Mo, Ba, and Be. In addition, the F1 proportions of most heavy metals in untreated MSWIFA were generally lower than those of F2 and F3, following the order of F3 (8%–27%) > F2 (1.4%–14%) > F1 (0.6%–14%) (Fig. 2a). For most heavy metals, the sequence was not changed after S/S treatment, i.e., F3 (9%–28%) > F2 (0.9%–16%) > F1 (0%–8%) (Fig. 2b). Fig. 2a illustrated the mild acid-soluble fractions of heavy metals in untreated MSWIFA followed the order of Cd (13%) > Mn (10%) > Co, Ba (9%) > Zn (6%) > Ni, Be (5%) > Mo, Hg, Cu, Sb, Pb, Cr, As (<3%). Cd, in medium risk rank, showed more mobile and bioavailable, which meant it was more hazardous if released into environment. S/S treatment significantly reduced the environmental risk of Cd and Mn, as well as some other heavy metals. In other words, SDD and cement showed high S/S efficiencies on most heavy metals, evaluated by F1 fractions (Table S2), wherein Cd (99.1%), Co (98.5%), Cu (98.1%), Ni/Mn (92.5%), etc.

The mobility and bioavailability of these fractions followed the sequence of F1 > F2 > F3 > F4 (Wang et al., 2015). Although heavy metals in reducible and oxidizable fractions were more stable than in mild acid-soluble fraction, they also could be released into environment if external conditions changed. Table S2 illustrated that S/S treatment reduced all heavy metals in the reducible fraction and oxidizable fractions, but to varying degrees. The significant decrease of Cd in mild acid-soluble fraction (i.e., from 14% to 0.3%) might increase the proportions of the other three fractions, which was also confirmed by a previous study (Wang et al., 2015). Another changing model happened on Pb, i.e., the decreasing reducible and oxidizable fractions increased the residual fraction. Overall, the sum of reducible and oxidizable fractions was decreased for most heavy metals. Hence, the S/S treatment process could achieve a good S/S effect on heavy metals by fractions transformation to reduce their mobility and bioavailability.

RAC method was used to evaluate the environmental risk of heavy metals in MSWIFA and the S/S treatment effect (Fig. 3). In untreated MSWIFA, the RAC values were distributed as three ranks, wherein medium risk (Cd (13.5%) > Mn (10.0%)), low risk (Co (8.7%) > Ba (8.5%) > Zn (5.5%) > Ni, Be (4.6%) > Mo (3.0%) > Hg (2.3%) > Cu, Sb (1.6%) > Pb (1.2%) > Cr (1.0%)), and no risk (As (0.6%)). The general risk ranks of the untreated MSWIFA were low compared with other 16 different sites in China, which reported that 53% of samples fell into high to very high-risk ranks due to Cd and Pb (37.5%–82.5%). (Pan et al., 2013; Wang et al., 2015). In this study, the control of Cd and Mn was still necessary before landfill, although they showed lower medium risk. S/S treatment downgraded the risk level of Cd, Co, Be, and Cu from medium/low risk to no risk, respectively. The Mn, Ba, Ni, and Pb also decreased by varying degrees but without downgrading the risk level. The Zn, Cr, Mo, As, Sb, and Hg were still at the low-risk rank. The increasing RAC values of Cr, Mo, and Hg could attribute to their diffusion, surface wash-off, and dissolution characteristics. Moreover, their low concentration and the cement addition could also result in these slight increases. Overall, the S/S formulation of SDD and cement could effectively control the environmental risk of heavy metals in MSWIFA by decreasing heavy metals in the mild acid-soluble fraction, i.e., reducing their mobility and bioavailability.

The above analysis confirmed the current S/S method, i.e., SDD (1%) + cement (10%), can well meet the emission control requirements of heavy metals and PCDD/Fs. However, 10% addition of cement will not only caused an unignorable volume increase but also increased the carbon dioxide footprint from the production process of cement, leading to the reduction of potential landfill capacity of MSWIFA and the environmental advantages. Hence, it will be better to design suitable and specific S/S methods for different sources of fly ash, including the choice of the appropriate additives, the optimum doses of reagents, etc. The MSWIFA with low environmental risk can be treated by SDD alone, maybe with a higher ratio of SDD. The MSWIFA with higher environmental risk was suggested to be treated by new chelate reagents with better S/S effect, such as SGA, TBA, EDTA, novel chemical polymers, etc. (Wang et al., 2015; Li et al., 2019; Ma et al., 2019a, b; Zhang et al., 2020).

3.2.3. *Heavy metals in liquid samples*

Table 3 illustrated the concentrations of heavy metals in leachate, river water, and groundwater, as well as their limitation in the MSWIFA leachate, which were detected according to the national standards (HJ 700-2014 and GB 7467-87) (MEEPRC, 1987, 2014). Their concentrations (mg/L) in leachate followed the order of Zn (0.9614) > As (0.0961) > Ni (0.0418) > Cu (0.0368) > Cr (0.138) > Cr $^{6+}$ (0.0117) > Ba (0.0112) > Pd (0.0104) > Se (0.0039) > Cd (0.0030), which were 1–4 orders of magnitude lower than the limited values in GB 16889-2008 (MEEPRC, 2008a). The leachate treatment process effectively reduced the concentrations of heavy metals by 94% (Zn), 80% (Cd), 77% (As), 62% (Se), 58% (Cr $^{6+}$), 58% (Cr), 39% (Pd), 29% (Cu), 23% (Ba), 1.0% (Ni), resulting in much lower concentration than national standard. It was attributed to the positive effect of coagulation/flocculation (Smaoui et al., 2016; Djeffal et al., 2021) and the adsorption/filtration process (Jaradat et al., 2021) on removing heavy metals in leachate.

In the river water and groundwater, the concentrations of some heavy metals (i.e., Cr, Cr $^{6+}$, Be, Ni, Pb, and Hg) were lower than the detection limit. The concentrations of most heavy metals decreased from upstream groundwater to downstream groundwater other than Ba and As. In addition, most of them had met the highest quality standards (I and II) in GB/T14848-2017, and only the Ni and As were in the III and Ⅳ

Table 3
The concentrations of heavy metals in liquid samples (mg/L).

	Zn	Pd	Cu	Cr	Ni	Cd	Cr^{6+}	Ba	As	Se	Be	Hg
L-R	0.9614 ±0.0056	0.0104 ±0.0018	0.0368 ±0.0015	0.0138 ±0.0011	0.0418 ±0.0013	0.0030 ±0.0007	0.0117 ±0.0011	0.0112 ±0.0019	0.0961 ±0.0013	0.0039 ±0.0006	ND	ND
L-T	0.0629 ±0.0034	0.0063 ±0.0009	0.0261 ±0.0018	0.0058 ±0.0008	0.0412 ±0.0012	0.0006 ±0.0002	0.0049 ±0.0007	0.0086 ±0.0009	0.0224 ±0.0011	0.0015 ±0.0004	ND	ND
GB 16889-2008	100	0.25	40	4.5	0.5	0.15	1.5	25	0.3	0.1	0.02	0.05
Ratio of L-T to Limited value	0.0006	0.0252	0.0007	0.0013	0.0824	0.0040	0.0033	0.0003	0.0747	0.0150		\
Removal efficiency (%)	94	39	29	58	1.4	80	58	23	77	62	\	\
RW	0.0126 ±0.0009	0.0001 ±0.00002	0.0009 ±0.0001	ND	ND	0.0002 ±0.00003	ND	0.0004 ±0.00008	0.0103 ±0.0012	0.0027 ±0.0005	ND	ND
GW-U	0.0502 ±0.0021	ND	0.0032 ±0.0006	ND	0.0158 ±0.0010	0.0003 ±0.00005	ND	0.0009 ±0.00007	0.0064 ±0.0003	0.0065 ±0.0002	ND	ND
GW-D	0.0057 ±0.0006	ND	0.0023 ±0.0004	ND	0.0055 ±0.0003	0.0002 ±0.00004	ND	0.0016 ±0.0002	0.0129 ±0.0013	0.0043 ±0.0008	ND	ND
GB/T14848-2017	0.5 (II)	0.005 (I)	0.01 (I)	/	0.02 (III)	0.001 (II)	0.005 (I)	0.01 (I)	0.05 (Ⅳ)	0.01 (I)	0.0001 (I)	0.0001 (I)
Detection limit (μg/L)	0.67	0.02	0.08	0.11	0.06	0.05	0.05	0.20	0.12	0.41	0.04	0.006

Notes: (I) and (II) represents the GW was suitable for various applications; (III) represents the GW was suitable for centralized domestic drinking water and industrial/agricultural water; (Ⅳ) represents the GW was suitable for industrial/agricultural water and centralized domestic drinking water after treatment.

standards, respectively (MNRPRC, 2017). For the river water, the concentrations of heavy metals were even lower than the groundwater requirements in GB/T14848-2017. All of which indicated the landfill disposal technology showed no influence on the surrounding groundwater and river water. The S/S treatment of MSWIFA and leachate treatment process provided adequate protection to the surrounding liquid media.

4. Conclusions

This study focused on the S/S effect on PCDD/Fs and heavy metals in the MSWIFA. Furthermore, their spatial distribution characteristics in other multiple media (soil, landfill leachate, groundwater, and river water) in the vicinity of the landfill were also investigated. S/S treatment significantly declined the mass/toxic concentrations of PCDD/Fs and heavy metals in MSWIFA due to the chemical fixation and dilution effect. BCR and RAC results further revealed the S/S mechanism of SDD and cement was decreasing heavy metals in the mild acid-soluble fraction to reduce their mobility and bioavailability. The oxidation treatment of leachate decreased the PCDD/F concentration by suspension absorption or NaClO oxidation decomposition, whereas a phenomenon of "memory effect" in the following processes (adsorption, press filtration, flocculating settling, slurry separation, and carbon filtration) increased the PCDD/F concentrations. Moreover, the leachate treatment effectively reduced the heavy metals to 1–4 orders of magnitude lower than the national emission standards.

Through the determination of PCDD/Fs and heavy metals in multiple media (landfill leachate, groundwater, and river water), no evidence showed any influence of the landfill on the surrounding liquid media. The slightly higher concentration of PCDD/Fs in the S-DW and S-NL could be influenced by other waste management processes (transportation and unloading) and other local sources beyond the landfill (e.g., hazardous incineration plant). The results indicated that proper control and management of landfills and leachate would not hazard the surrounding environment, and the MSWIFA S/S treatment and multi-step leachate treatment can be extended to more landfills. However, the sampling numbers were not enough to conduct complex statistical analysis to better assess the environmental impact, which should be improved in future work.

Author statement

Xiaoqing Lin: Conceptualization, Methodology, Writing - Review & Editing. **Yunfeng Ma**: Data curation, Writing - Original Draft, Writing - Review & Editing. **Tong Chen**: Data curation, Funding acquisition. **Lei Wang**: Conceptualization, Writing - Review & Editing. **Masaki Takaoka**: Writing - Review & Editing. **Shuping Pan**: Conceptualization, Resources. **Hao Zhang**: Software, Writing - Review & Editing. **Angjian Wu**: Visualization, Investigation. **Xiaodong Li**: Supervision, Conceptualization. **Jianhua Yan**: Supervision, Project administration.

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.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

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