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Chlorobenzenes and PCDD/Fs emissions from a pilot-scale plasma melting incinerator of simulated radioactive waste: Impacts of waste composition and operational parameters

Wenqian Jiang [a], Lulu Dong [a], Minghui Tang [a,b,*], Shengyong Lu [a,**], Fanjie Shang [c], Jie Lin [c], Shaofu Tang [c]

[a] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, 310027, PR China
[b] Qingshanhu Energy Research Center Zhejiang University, 1699 Dayuan Road, Qingshanhu Science and Technology City, Hangzhou, 311305, PR China
[c] Zhejiang Fuchunjiang Environmental Technology Research Co., Ltd., Hangzhou, 311401, PR China

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ABSTRACT

The efficient and safe disposal of radioactive waste remains a critical global challenge. While plasma technology is widely applied for its efficiency and safety, studies on secondary pollutant emissions remain limited. This research investigated emissions of chlorobenzenes (CBzs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) during the disposal of simulated radioactive waste from a pilot-scale high-temperature plasma melting incinerator in China under different waste conditions with rubber, polyvinyl chloride (PVC), and waste resin. Total concentrations of CBzs and PCDD/Fs in stack gas ranged from 3.445 to 117.192 μg/Nm^3 and 0.006–0.408 ng TEQ/Nm^3, respectively. High-chlorine wastes such as waste resin, PVC, and rubber increase the emissions of CBzs and PCDD/Fs, while co-adding PVC and rubber reduces PCDD/Fs emissions due to synergistic effect. Dominant congeners included 2,3,7,8-TCDD, 2,3,4,7,8-PeCDF, and HCBz. Furthermore, correlation analysis exhibited a markedly linear correlation between 1,4-DCBz and PCDD/Fs, and the differential pressure between the quench tower and filter correlated positively with PCDD/Fs, particular when PVC and rubber were added. This research not only explores the impact of waste type and parameters of air pollution control devices on PCDD/Fs emissions, but also provides significant data to enhance the safe disposal of radioactive waste and improves plasma technology processes.

1. Introduction

As the nuclear industry continues to advance, the generation of radioactive waste has become an inevitable outcome of nuclear power plants, medical applications, scientific research institutions, and industrial activities [1]. The disposal of radioactive waste is a major challenge faced worldwide. Nuclear power plants around the world produce thousands of tons of high-, medium-, and low-level waste each year. Among them, low-level radioactive waste, which includes items such as contaminated protective clothing, tools, and cleaning materials with relatively low radiation levels, comprises over 95 % of the total volume of radioactive waste [2,3]. The growth in radioactive waste will cause potential risks to both ecological systems and human health. According to statistics in China, nuclear power operations generated 1255 m^3 of new radioactive waste in 2022, and the radioactive waste temporarily stored in nuclear power plants reached 14,691 m^3 [4]. Therefore, exploring effective methods and technical status for disposing of radioactive waste holds great strategic significance.

Plasma technology has been recognized by the International Atomic Energy Agency as an effective technical method for disposing low-level radioactive waste [5,6] due to its high efficiency, safety, and resource recovery characteristics. Switzerland, Russia [7], Bulgaria [8], and other countries already have large-scale plasma melting furnaces for the disposal of low-level radioactive waste [9]. The application of plasma technology for radioactive waste disposal in China started relatively late. Some research institutes have conducted certain experimental studies on waste resins, polyethylene (PE), polyvinyl chloride (PVC), and other wastes [10–13]. The research focus is more on the

* Corresponding author. State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, 310027, PR China.
** Corresponding author.
E-mail addresses: lytmh1214@zju.edu.cn (M. Tang), lushy@zju.edu.cn (S. Lu).

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solidification characteristics of radionuclides, and there is less research on the secondary pollutant emissions generated during the disposal process of this technology.

Plasma technology can effectively destroy organic pollutants and reduce the formation of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) due to its high incineration efficiency and thorough incineration effect [13]. It is considered that it can achieve low-level emissions of conventional pollutants like SO_2 and NO_x, as well as persistent organic pollutants (POPs) like PCDD/Fs in the flue gas during radioactive waste disposal. Relevant studies have shown that the amount of waste generated and discharged into the atmosphere by plasma treatment is 1.5–2 times lower than that of incineration treatment [14]. At the same time, plasma technology has a significant effect on reducing and decomposing harmful pollutants. Yoshida et al. [15] used pulsed power plasma technology to study the simultaneous removal of NO_x and PCDD/Fs. The results indicated that when the gas flow rate reached 5000 Nm^3/h, 75–84 % of PCDD/Fs could be decomposed and destroyed, and 93 % of NO was converted into NO_2, which significantly reduced the emission of harmful gases during the treatment process. Domestically, Yan et al. [16] used sliding arc plasma to treat PCDD/Fs in simulated flue gas and also achieved 30 %–70 % removal of PCDD/Fs I-TEQ values. In addition, Pourali [17] conducted a study on the plasma pyrolysis treatment system of radioactive hazardous waste generated by a floating nuclear power plant in northern Russia [14] and found that the PCDD/Fs emissions in the flue gas produced by the system was 0.014–0.02 ng TEQ/Nm^3. This result is lower than the PCDD/Fs emission of pyrolysis and incineration of radioactive waste in previous studies with an average of 0.023 ng TEQ/Nm^3 [18], indicating that although plasma technology can effectively decompose and destroy PCDD/Fs, the PCDD/Fs produced during the plasma disposal of radioactive waste cannot be ignored.

Waste composition is considered to be one of the key factors affecting pollutant emissions and the distribution of PCDD/Fs congeners during incineration [19,20]. The team of Bobrakov, A. N. studied the influence of the type of simulated low-level radioactive waste disposed on pollutant emissions in a vertical furnace plasma system at the Russian Center for Demonstration of Radioactive Waste Management Technologies [21]. The results showed that when disposing of high-ion exchange resin, the HCl and SO_x concentrations in the extracted dry pyrolysis gas reached 100 mg/m^3 and 10,000 mg/m^3, respectively, which is much higher than the HCl (26 mg/m^3) and SO_x (50 mg/m^3) concentrations in the pyrolysis gas when disposing of high-polyethylene (PE) waste. During the plasma melting process of mixed radioactive waste [7], the HCl and SO_x concentrations in the exhaust flue gas were detected to be 0.015 mg/m^3 and 0.050 mg/m^3, respectively, and PCDD/Fs were detected in the flue gas with 0.004 ng TEQ/Nm^3. It can be seen that different types of radioactive waste have different pollutant emission characteristics after being treated with plasma technology. Therefore, while exploring the emission characteristics of PCDD/Fs and other pollutants during the plasma treatment of radioactive waste from nuclear power plants, it is also urgent to conduct more in-depth research on the types and compatibility of radioactive waste.

It can be seen that plasma technology is currently used to treat radioactive waste around the world [9]. However, the current research focus is more on the volume reduction and reduction characteristics of waste [11,12,22–24]. Existing studies still lack sufficient research on the emission characteristics of stack flue gas pollutants, especially POPs such as PCDD/Fs, during the disposal of radioactive waste. Although plasma technology can effectively destroy organic matter and reduce pollutant emissions, the pollutant emission problem of plasma technology in treating nuclear power plant radioactive waste cannot be completely ignored, especially the emission of PCDD/Fs, which are among the most toxic and persistent organic pollutants known to date.

This study explored the emission of PCDD/Fs and chlorobenzenes (CBzs) during the disposal of simulated radioactive waste from a nuclear power plant in a high-temperature plasma melting device in China. The effects of different waste types, the addition of high-chlorine waste in the materials, and air pollution control devices (APCDs) parameters on the emission of CBzs and PCDD/Fs were studied. In addition, the relationships between PCDD/Fs, operating parameters, and conventional pollutants under various waste type conditions were explored based on the results of Pearson correlation coefficient, principal component analysis (PCA), and cluster analysis. This study hopes to provide data support for the safe disposal and environmental friendliness of radioactive waste in the future, and achieve wider application and development. Carrying out these studies in high-temperature plasma melting devices will help achieve low-level emissions of PCDD/Fs in the future disposal of radioactive waste from nuclear power plants and better achieve emission reduction.

2. Materials and methods

2.1. Experimental design

This study was carried out from a plasma high-temperature melting furnace in the southwest of China. The Average concentrations of traditional pollutants and operating parameters of the plasma melting furnace are shown in Table 1. The system is an engineering prototype with a practical average daily disposal capacity of 300 kg/day. The average melting temperature of the first combustion chamber is 840 °C, and the temperature of the flue gas entering the air pollution control device (APCDs) after passing through the second combustion chamber is 833 °C. The off-gas quenching system rapidly cools the flue gas from 163 °C to 74 °C.

The waste entering the melting furnace is simulated radioactive nuclear waste mixed with vitrified formula additives, including dry wastes such as PE, cotton, paper clothes, rubber, PVC, and wet wastes such as waste resin. These materials are selected to simulate the actual types of radioactive waste generated during the disposal of a nuclear power plant, including protective clothing (represented by cotton and paper clothes) that can become contaminated, polymeric materials (PE, PVC, rubber), and ion exchange resins (waste resin) commonly used in reactor coolant systems [10,13]. The APCDs in this system are composed of a quenching tower, a dust collector, an absorption tower, a mist eliminator, and a carbon bed combined adsorber, as shown in Fig. 1. Samples were collected at the chimney over a period of twelve days.

To investigate how the type of waste fed into the furnace impacts the emission of PCDD/Fs and CBzs, six combinations of three types of waste were analyzed, including: (1) wet waste (WW): waste resin; (2) miscellaneous dry waste (DW): PE, cotton, paper clothing, rubber, PVC; (3) mixed waste (MW): a mixture of waste resin and dry wastes such as PE and cotton. The detailed sampling arrangements are presented in Table 2. Among them, the total chlorine content of the combustible part of the samples obtained by oxygen bomb combustion-ion chromatography as shown in Table 3 shows that PVC and rubber in miscellaneous

Table 1
Average concentrations of traditional pollutants and operating parameters of the plasma melting furnace.

Parameters	
Design capacity (t/d)	1.2
Actual average capacity (t/d)	0.3
Melting temperature of combustion chamber (°C)	840.1
Temperature of second combustion chamber outlet (°C)	833.3
Temperature of quench tower inlet (°C)	162.8
Temperature of quench tower outlet (°C)	74.4
Temperature of stack flue gas (°C)	39.0
O_2 (%)	15.6
SO_2 (mg/Nm^3)	31.6
NO_x (mg/Nm^3)	512.0
CO (mg/Nm^3)	92.3
HCl (mg/Nm^3)	2.8
Particulate matter (mg/Nm^3)	46.0

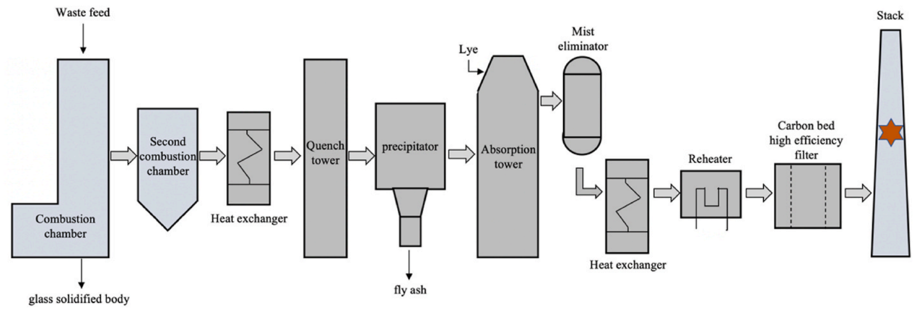

Fig. 1. The systemic diagram and sampling point of the plasma melting furnace.

Table 2
Detailed information on the disposal of simulated radioactive nuclear waste in a plasma melting furnace.

Waste Type	Waste composition	Average feed quantity (kg/h)	Sample number
Wet waste (WW)	Waste resin	27	1–2
Miscellaneous dry waste (DW)	DW1: Pure miscellaneous dry waste[a]	33	3–8
	DW2:DW1+Rubber[b]	38	9–11
	DW3:DW1+PVC[c]	23	12–21
	DW4:DW1+Rubber + PVC[d]	36	22–24
Mixed waste (MW)	DW1+Waste resin[e]	32	25–32

[a] DW1: (50 % PE, 50 % cotton) or (50 % PE, 50 % paper clothing) or (30 % PE, 30 % cotton, 50 % paper clothing).
[b] DW2: (47.9 % cotton, 47.9 % PE, 4.2 % rubber).
[c] DW3: (61.2 %PE, 30.6 % cotton, 8.2 %PVC).
[d] DW4: (45.5 %PE, 30.3 % cotton, 20.2 paper clothing, 2.0 % rubber, 2.0 % PVC).
[e] MX: (42.3 %PE, 21.1 % cotton, 36.6 % waste resin).

Table 3
Results of determination of chlorine content in the furnace waste.

Waste Type	Cotton	PE	Paper clothing	Rubber	PVC	Waste resin
Cl (%)	0.03	0.15	0.2	0.29	3.27	0.58

dry waste have relatively high chlorine contents of 3.27 % and 0.29 %, respectively. Therefore, miscellaneous dry waste was divided into four types according to whether it contained rubber and PVC, and studied separately to further explore the impact of waste type entering the furnace on PCDD/Fs and CBzs emissions. The selection of these types allows not only to explore the effects of different chlorine content gradients, but the mixed disposal of wastes also breaks through the limitations of a single material and more closely matches the needs of practical co-disposal of multiple wastes.

2.2. Sampling, analyzing and QA/QC

When operating conditions were stable, thirty-two PCDD/Fs and CBzs samples were collected directly from the stack gas using isokinetic samplers (ZR-3720, Junray, China) in accordance with the EPA method 23a [25]. In order to ensure the accuracy and reliability of the findings, at least two samples were tested in parallel with an average collection time of 1.5 h. All collected samples were pretreated for PCDD/Fs within two weeks after being collected on-site. After pretreatment, extraction, and purification, the concentrations of separated PCDD/Fs congeners in the purified samples were analyzed using high-resolution gas chromatography with high-resolution mass spectrometry (HRGC/HRMS) (JMS-800D, JEOL, Japan, DB-5MS silica capillary column, 60 m × 0.25 mm × 0.25 μm). The pretreatment and analytical protocols for PCDD/Fs were adapted based on previously reported methods [26,27] and further refined for the specific conditions of this study. Quality control measures ensured the average recoveries of PCDD/Fs standards ranged from 38.4 % to 156 %, meeting the EPA Method 23a criteria.

For the analysis of CBzs, the purified samples were analyzed using gas chromatography coupled with mass selective detection (GC/MSD) (Agilent 6890N GC/5975 B MSD, DB-5MS column, 30 m × 0.25 mm × 0.25 μm). The compete CBzs method was detailed described in previous studies [28]. During the experiment, all samples were stored in half to ensure a qualified recovery rate. To ensure comparability, all reported concentrations were standardized to standard condition where oxygen content was 11 %. In addition, to study the impact of different types of waste entering the furnace on CBzs emissions and the correlation between CBzs and PCDD/Fs, CBzs emissions in flue gas samples were measured at the same time. The process parameters of plasma melting disposal, the operating parameters of APCDs, and conventional flue gas pollutants at the tail were obtained synchronously by the on-site Continuous Emission Monitoring System (CEMS) online detection system.

2.3. Statistical analysis

In this study, the I-TEQ values for PCDD/Fs were calculated using the international toxic equivalent factors (I-TEF), in accordance with the standard for pollution control on the municipal solid waste incineration (MSWI) (GB18485-2014) and hazardous waste incineration (HWI) (GB18484-2020) in China. The total I-TEQ values of the 17 toxic 2,3,7,8-substituted PCDD/Fs congeners were calculated as follows and the concentrations are provided in Table S2:

$$C_{TEQ} = \sum C_i \times TEQ_i \quad (1)$$

C_i represents the concentration of each toxic PCDD/Fs congener; TEQ_i represents the TEQ values of each toxic PCDD/Fs congener; C_{TEQ} represents the total I-TEQ values of the sample.

To explore the relationships among CBzs, traditional pollutants, operating parameters, and PCDD/Fs congeners, statistical analyses including Pearson correlation coefficient (r), PCA, and hierarchical clustering were conducted using SPSS 24.0 software. The Pearson correlation was used to measure linear correlations between variables, where PCA transformed the original variables into independent comprehensive indices, highlighting their correlations. Hierarchical clustering provided a bottom-up approach to reveal the hierarchical structure among variables.

3. Results and discussion

3.1. PCDD/Fs and CBzs concentrations under different waste types conditions

CBzs and PCDD/Fs were analyzed to investigate the emission characteristics of chlorinated organic pollutants in different simulated nuclear power plant wastes. Table 4 summarized the CBzs and PCDD/Fs emissions in the stack gas of thirty-two samples from the plasma melting furnace under different waste types conditions. To examine the impact of waste type on CBzs and PCDD/Fs emissions, their concentrations are illustrated in Fig. 2(a)(b)(d). With the aim of exploring the effect of waste type on the chlorination degree of PCDD/Fs, the chlorination degree and the PCDDs/PCDFs ratio are presented in Fig. 2(c).

The I-TEQ values of PCDD/Fs of the stack gas ranged from 0.006 to 0.408 ng TEQ/Nm^3 (0.531–19.942 ng/Nm^3) as shown in Table 4. All PCDD/Fs emissions were below the standard for pollution control on HWI (0.5 ng TEQ/Nm^3) (GB 18484–2020), and 50 % of samples exceeded the MSWI standard (0.1 ng TEQ/Nm^3) outline in GB 18485–2014. This is because the platform is a pilot platform and the emission characteristics of wastes with different compositions are still in the research stage. The concentrations of CBzs (3.445–117.192 μg/Nm^3) were significantly higher than those of PCDD/Fs, aligning with previous studies in MSWIs [29] and HWIs [27,28]. Furthermore, a similar trend was observed between CBzs and PCDD/Fs concentrations in response to variations in waste types.

Due to changes in the compatibility and type of materials at the front end during the operation of the plasma melting prototype, there are differences in dioxin emissions during the incineration process of different materials. It can be seen from the display in Fig. 2 that among the six waste type conditions, the combination of pure miscellaneous dry waste and rubber (DW2) has the highest emissions of CBzs and PCDD/Fs, which are 81.981 μg/Nm^3 and 0.311 ng TEQ/Nm^3 respectively. Compared with the pure miscellaneous dry waste (DW1) condition, the possible reasons for the high emissions are not only the certain concentration of chlorine (0.29 %) in the rubber but also the maximum average feed amount. Also, it may be that the average melting temperature of the combustion chamber in DW2 condition is only about 700.9 °C, and the combustion temperature is too low to fully decompose chlorine-containing compounds, thus forming more dioxins (Table S1). Specifically, the formation of PCDDs under this working condition is much greater than the formation of PCDFs, and the ratio of PCDDs/PCDFs reaches 2.01. The results show that the formation pathway of PCDDs is more influenced by the chlorine content of waste and melting temperature of the furnace compared to PCDFs, aligning with previous findings in MSWI [30].

In other miscellaneous dry waste (DW) conditions, PVC is also added to the furnace as a type of dry waste. Although PVC has the highest chlorine content with 3.27 %, the combination of miscellaneous dry waste and PVC (DW3) has an average I-TEQ value of 0.031 ng TEQ/Nm^3, which is lower than the PCDD/Fs concentration (0.111 ng TEQ/Nm^3) of DW1. However, the CBzs concentration of DW3 (24.548 μg/Nm^3) is higher than that of DW1 (15.032 μg/Nm^3). The possible reason is that the particulate matter concentration of stack gas is controlled at a relatively low level under the DW3 operating condition (Table S1), indicating that more solid-phase PCDD/Fs attached to the particulate matter is removed in the APCDs, resulting in lower levels of PCDD/Fs emissions. At the same time, the PVC doped in DW3 decomposes into CBzs at a certain temperature, but the melting temperature of the first combustion chamber (Table S1) of 769.4 ° C and the residence time under this working condition may not be conducive to the formation of PCDD/Fs. For the combination of miscellaneous dry waste, rubber, and PVC (DW4), due to the combination of rubber and PVC, two wastes with high chlorine content, the emissions of CBzs and PCDD/Fs reached 47.875 μg/Nm^3 and 0.200 ng TEQ/Nm^3 respectively. At the same time, the ratios of PCDDs/PCDFs under conditions DW3 and DW4 were 0.78 and 0.10, respectively (Fig. 2(c)). This result indicates that the addition of PVC to the waste entering the furnace may cause the formation of PCDFs to be higher than PCDDs, which is consistent with previous studies on the use of plasma arc melter system to treat typical mixtures of low-level radioactive waste [13].

The CBzs and PCDD/Fs emissions for the mixed waste condition (MW), consisting of waste resin and pure miscellaneous dry waste, were 38.830 μg/Nm^3 and 0.156 ng TEQ/Nm^3, respectively. These emission concentrations were higher than the concentrations of the WW condition with only waste resin and DW1 condition with only pure miscellaneous dry waste. Although WW was mixed with pure miscellaneous dry waste, the PCDDs/PCDFs ratios of WW and MW conditions were close, both close to 1.53. This result shows that the addition of pure miscellaneous dry wastes such as PE, paper clothes, and cotton to waste resin has little effect on the proportion of PCDDs and PCDFs in the tail flue gas. From the above results, it can be seen that the selection of the feeding waste compatibility and type is particularly important during the operation of the plasma melting prototype. It is necessary to select the appropriate waste compatibility according to the pyrolysis and incineration characteristics of different wastes, so that the pollutant emissions of stack gas can reach a cleaner and more efficient level.

Table 4
The summary of the measurements for pollutants in stack gas.

Waste type	Sample number	CBzs (μg/Nm^3)	PCDDs (ng/Nm^3)	PCDFs (ng/Nm^3)	PCDD/Fs I-TEQ (ng TEQ/Nm^3)
WW	1	39.801	4.882	2.336	0.109
	2	26.579	1.388	1.417	0.042
	average	33.190	3.135	1.877	0.075
DW1	3	25.354	2.446	3.625	0.071
	4	17.321	2.192	8.300	0.072
	5	6.144	1.262	6.774	0.104
	6	3.445	1.117	4.642	0.060
	7	18.541	1.148	5.430	0.072
	8	19.385	1.703	16.755	0.285
	average	15.032	1.645	7.588	0.111
DW2	9	58.462	14.807	5.135	0.408
	10	70.287	11.765	4.396	0.260
	11	117.192	4.783	10.118	0.265
	average	81.981	10.452	6.550	0.311
DW3	12	34.971	6.613	1.569	0.102
	13	16.358	0.581	0.888	0.021
	14	14.024	0.146	0.386	0.006
	15	15.821	0.256	0.795	0.012
	16	14.739	0.149	0.542	0.008
	17	25.954	1.204	8.551	0.072
	18	40.213	0.619	0.670	0.008
	19	42.514	0.238	0.667	0.008
	20	22.079	0.421	1.076	0.014
	21	18.811	1.050	8.111	0.056
	average	24.548	1.128	2.325	0.031
DW4	22	25.802	1.476	13.542	0.212
	23	58.955	0.765	6.952	0.088
	24	58.868	1.336	16.935	0.300
	average	47.875	1.192	12.476	0.200
MW	25	46.461	7.604	5.975	0.164
	26	41.760	4.614	3.611	0.134
	27	49.771	7.517	3.246	0.123
	28	47.444	10.061	3.137	0.198
	29	36.975	9.123	3.400	0.161
	30	37.097	4.049	7.414	0.141
	31	33.247	5.669	7.316	0.224
	32	17.885	1.150	6.307	0.098
	average	38.830	6.223	5.051	0.156

3.2. PCDD/Fs and CBzs congeners distribution

3.2.1. Congeners profiles of PCDD/Fs

As shown in Fig. 3(b)(c), the fingerprints of 2,3,7,8-substituted and 136 PCDD/Fs congeners in stack gas under six waste types conditions are presented to elucidate the formation [31,32] and changing characteristics. TCDD and TCDF concentrations dominated the total distribution of the 136 congeners, followed by PeCDD and PeCDF. This result aligns with findings from the previous study in a rotary kiln medical

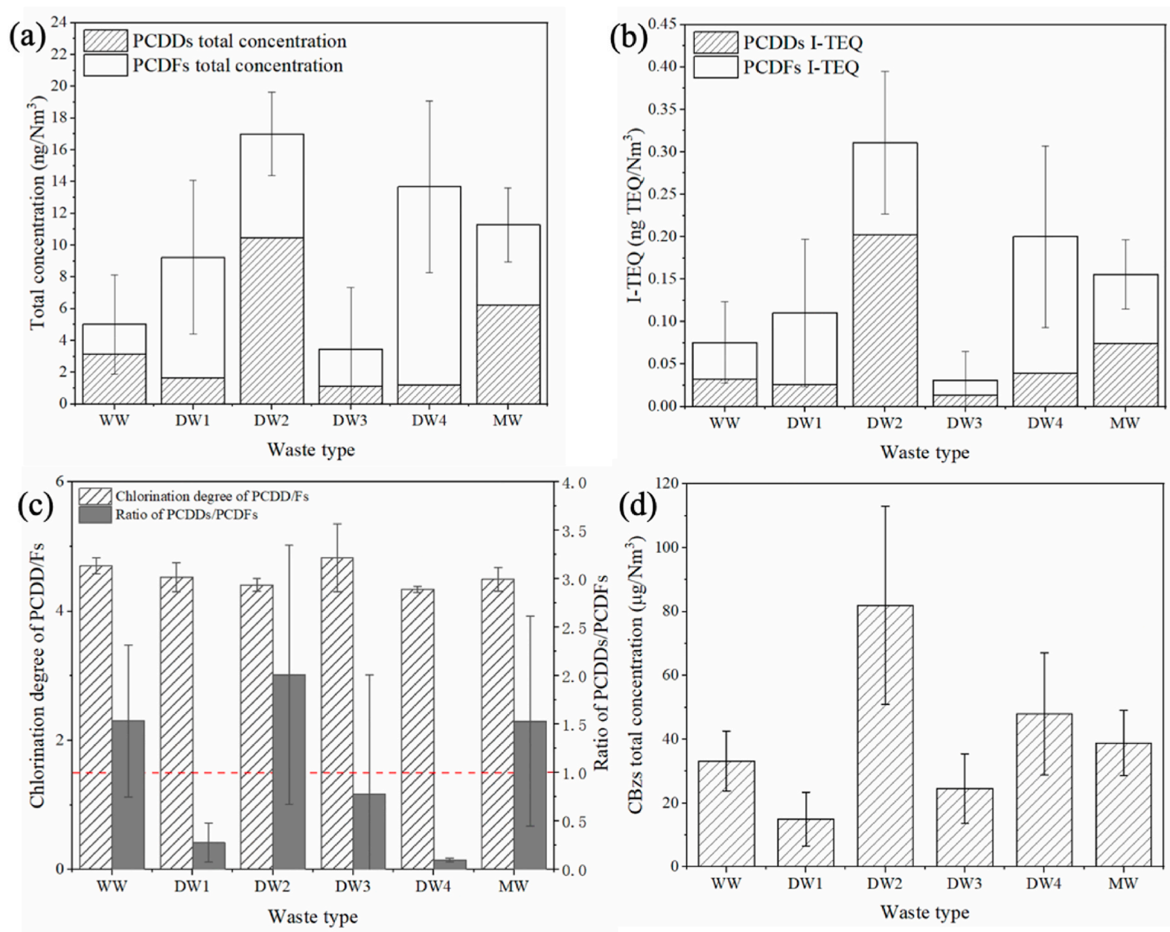

Fig. 2. (a) PCDDs and PCDFs concentrations; (b) PCDDs and PCDFs I-TEQ values; (c) chlorination degree of PCDD/Fs and ratio of PCDDs/PCDFs; (d) CBzs concentrations under six waste types conditions.

waste incinerator [33], but differs from those findings in MSWIs [34,35]. Notably, the concentrations of low-chlorinated PCDD/Fs were much higher than those of high-chlorinated PCDD/Fs. Specifically, TCDD is dominant in conditions containing wet waste (WW, MW) and DW2, accounting for 40.6 %–49.7 %. In conditions containing miscellaneous dry waste (DW1, DW3, DW4) other than DW2, TCDF accounts for the highest proportion, which takes approximately 47.4 %–70.6 %.

As shown in Fig. 3(b), the distribution of 2,3,7,8-substituted PCDD/Fs congeners in the reaction products differed according to the waste types. The 2,3,7,8-substituted PCDFs accounted for the majority (53.2 %–80.3 %) of the 17 toxic PCDD/Fs across all conditions except DW2, which is consistent with the previous findings in MSWIs [34] and HWIs [27,36]. In DW2 condition, 2,3,7,8-TCDD accounts for the highest proportion, which takes 54.1 %. The low-chlorinated PCDD/Fs concentrations (2,3,7,8-TCDD, 2,3,7,8-TCDF, and 2,3,4,7,8-PeCDF) dominate the distribution of the 17 PCDD/Fs congeners in all the operating conditions, which is in agreement with previous research results on plasma treatment of low-level radioactive waste [13]. In the conditions containing wet waste (WW, MW) and DW2, 2,3,7,8-TCDD is dominant, accounting for 34.9 %–54.1 %. In the conditions containing miscellaneous dry waste (DW1, DW3, DW4) except DW2, 2,3,4,7,8-PeCDF accounts for accounting for 27.0 %–47.2 %, which is the highest proportion. This result is similar to the characteristics of the 136 PCDD/Fs congeners mentioned above.

In the comparison of DW1, DW2, and DW3 conditions, the addition of rubber and PVC to the feed waste separately led to an increased proportion of PCDDs in the total PCDD/Fs concentrations as shown in Fig. 3(c). Differences in the PCDFs congeners profile were evident, with increased proportions of HpCDF and OCDF and decreased proportions of TCDF and PeCDF. This suggests that the chlorination reaction of PCDFs was enhanced as the chlorine content in the feeding waste increased. The most obvious change is the proportion of TCDF, which decreased by 33.9 % and 13.1 % respectively compared with DW1. A notable rise in the proportion of TCDD was observed, reaching the highest levels in the total PCDD/Fs concentrations, increased from 8.8 % to 49.7 % and 17.2 %. This result suggested that adding rubber and PVC to the feeding waste separately exerted a stronger enhancing effect on the formation of low-chlorinated PCDD/Fs than high-chlorinated ones. Among the distribution of 2,3,7,8-substituted PCDD/Fs congeners in Fig. 3(b), the proportions of 2,3,7,8-TCDD and 2,3,7,8-TCDF increased, and 2,3,4,7,8-PeCDF decreased. The remaining 2,3,7,8-substituted PCDD/Fs congeners are less than 10 % with little change.

However, it was found in the study that adding rubber and PVC (DW4) to miscellaneous dry waste at the same time has different PCDD/Fs emission characteristics than adding them separately. Studies have shown that the synergistic effect of rubber and PVC may lead to more PCDD/Fs production. Specifically, the proportion of PCDDs decreased, while the proportions of TCDF and PeCDF increased. Among the 2,3,7,8-substituted PCDD/Fs, 2,3,7,8-TCDD, 2,3,7,8-TCDF, and 2,3,4,7,8-PeCDF all have varying degrees of rise. These results are completely opposite to the PCDD/Fs distribution characteristics of the conditions where rubber and PVC were added alone (DW2, DW3). The possible reason for this result is that the organic matter in the rubber may react with the chlorine released from the PVC to generate more PCDFs.

For the condition containing wet waste (WW, MW), compared with DW1 condition, it has a higher proportion of TCDD and PeCDD and a

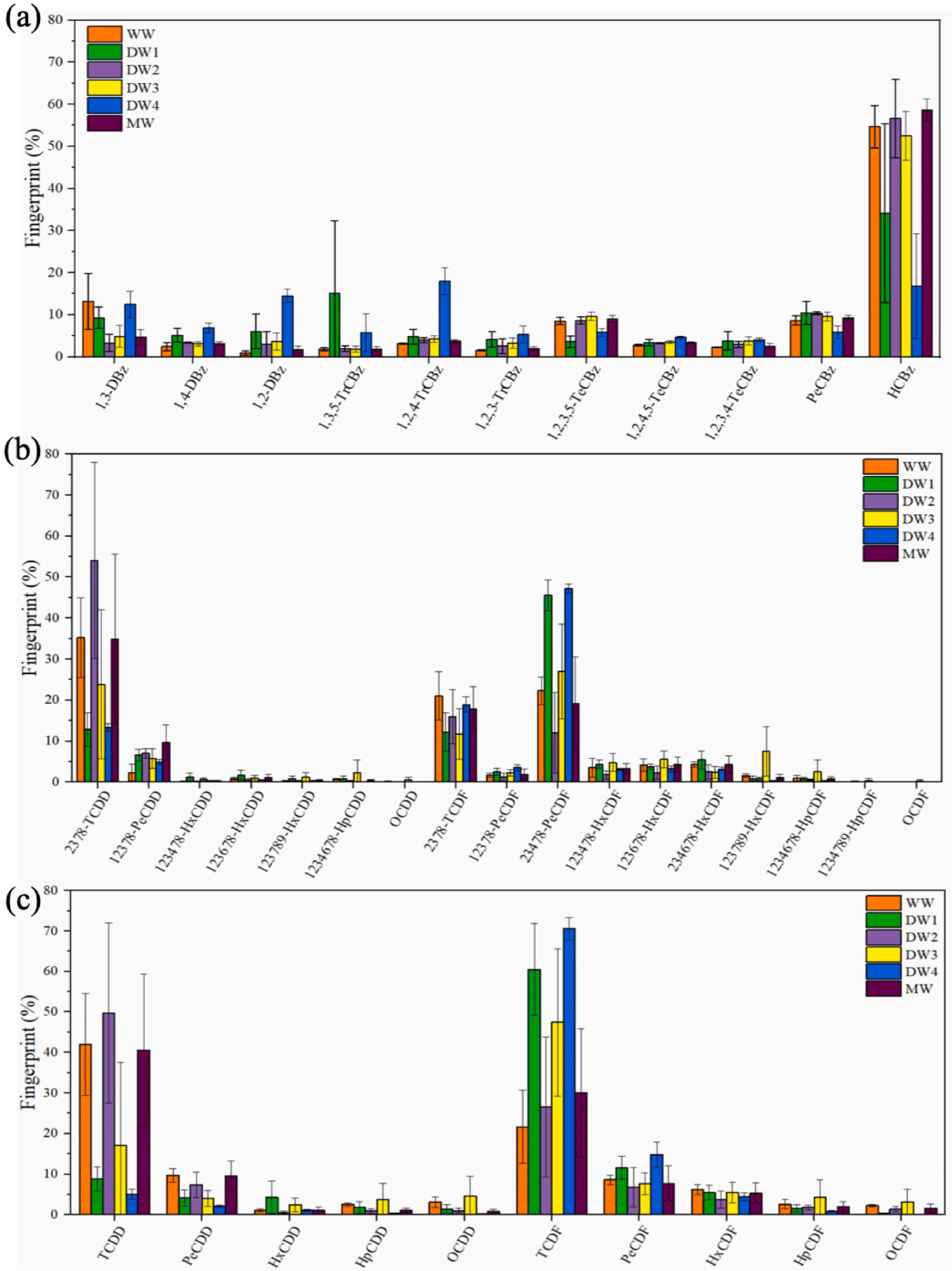

Fig. 3. Fingerprints of (a) PCDD/Fs congeners; (b) 2,3,7,8-substituted PCDD/Fs congeners; (c) CBzs congeners in stack gas under six waste types conditions.

lower proportion of TCDF and PeCDF. At the same time, after adding miscellaneous dry waste to the wet waste in MW condition, the proportion of TCDF increases and the proportion of PeCDF decreases. The proportions of other congeners remain almost at the same level. Among 2,3,7,8-substituted PCDD/Fs, the proportion 1,2,3,7,8-PeCDD increased in MW compared with WW condition, while the proportion of 2,3,7,8-TCDF and 2,3,4,7,8-PeCDF decreased.

3.2.2. *Congeners profiles of CBzs*

The congener profiles of CBzs are illustrated in Fig. 3(a) and detailed in Table S3. As depicted in Fig. 3(a), Hexachlorobenzene (HCBz) exhibited the highest concentration among the CBzs, which takes approximately 34.2 %–58.6 % of the total CBzs concentrations across all operating conditions, with the exception of DW4 condition. Pentachlorobenzene (PeCBz) and 1,3-dichlorobenzene (1,3-DCBz) followed in terms of concentration levels. This outcome was noticeably different from the results reported in the previous study conducted on a rotary kiln HWI [27], in which 1,3-DCBz was dominant. In DW4 condition, 1,2,4-trichlorobenzene (1,2,4-TrCBz) and HCBz have similar proportions and both dominant positions. In the WW, DW2, and MW conditions shown in Table S3, the HCBz concentration exceeded 15 μg/Nm^3, accounting for more than 50 % of the total CBzs concentration.

In the comparison of DW1, DW2, and DW3 conditions, the addition of rubber and PVC to the feeding waste separately led to a decrease in the contribution of low-chlorinated CBzs, such as DCBz and TrCBz. Conversely, the concentrations of high-chlorinated CBzs like 1,2,3,5-tetrachlorobenzene (1,2,3,5-TeCBz) and HCBz increased. These findings suggested that the separate addition of rubber and PVC more strongly promoted the formation of high-chlorinated CBzs compared to low-chlorinated ones. However, when rubber and PVC (DW4) were added to the miscellaneous dry waste, although the concentration of CBzs congeners increased, the fingerprint characteristics of CBzs changed in the opposite way. Compared with DW1, the proportion of high-chlorinated CBzs decreased, among which the proportion of HCBz decreased from 34.2 % to 16.8 %. The proportion of low-chlorinated CBzs increased, among which the proportion of 1,2,4-TrCBz increased the most, from 4.8 % to 18.0 %. The possible reason for this result is the synergistic effect of rubber and PVC [37]. The decomposition products of organic matter in rubber may react with the chlorine released in PVC to produce more low-chlorinated CBzs.

Due to the low chlorine content in miscellaneous dry waste, the emission concentration of CBzs did not change much after adding miscellaneous dry waste (MW) to wet waste (WW), which were 38.83 μg/Nm^3 and 33.19 μg/Nm^3 respectively. It is more obvious that when miscellaneous dry waste is added to wet waste, the proportion of 1,3-DCBz decreases from 13.2 % to 4.7 %, and the proportion of HCBz increases from 54.7 % to 58.6 %. Other CBzs congeners changed little. Compared with DW1 condition, both WW and MW conditions exhibited higher CBzs emission concentrations due to the presence of waste resin. The results highlighted that CBzs emissions are closely influenced by the composition and types of waste materials. Clarifying the different characteristics of material compatibility can provide valuable insights for reducing CBzs emissions effectively.

3.3. Correlation among CBzs, PCDD/Fs, traditional pollutants concentrations, and operational parameters

3.3.1. Multivariate analysis

To explore the relationships among CBzs, PCDD/Fs, traditional pollutants concentrations, and operational parameters under six waste-type conditions, PCA method was applied to analyze (Fig. 4(a)(c)). As depicted in Fig. 4(a), the sum of the two first factors is 62.1 % in the analysis of CBzs congener concentrations and toxic PCDD/Fs. The PCA results showed that strong correlations among toxic PCDD/Fs congeners, excluding 2,3,7,8-TCDD. All variables were positioned within the positive range of Factor 1 and Factor 2, indicating a positive correlation between CBzs and 17 toxic PCDD/Fs congeners. Meanwhile, Factor 2 shows a markedly positive correlation with 2,3,7,8-substituted PCDD/Fs congeners, especially high-chlorinated PCDD/Fs. In contrast, it exhibited a markedly negative correlation with high-chlorinated CBzs, including DCBz and TrCBz. Specifically, 1,2-DCBz, 1,3-DCBz, 1,4-DCBz,

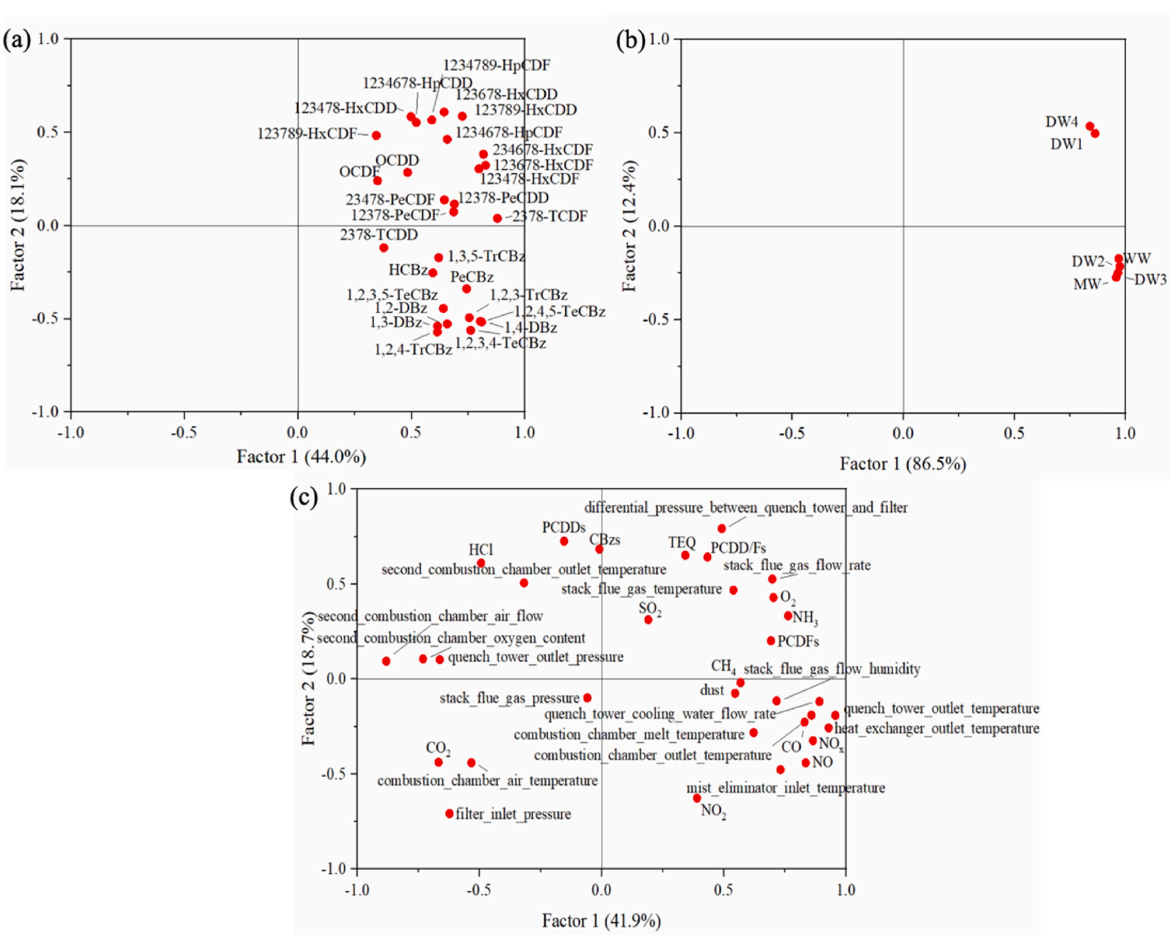

Fig. 4. The PCA analysis of (a) congener concentrations of CBzs and toxic PCDD/Fs; (b) waste types; (c) CBzs, PCDD/Fs, traditional pollution concentrations, and operational parameters.

1,2,3-TrCBz, 1,2,4-TrCBz, 1,2,3,4-TeCBz, and 1,2,4,5-TeCBz were positioned far from toxic PCDD/Fs congeners, suggesting a weak correlation with PCDD/Fs. However, strong correlations were observed between 1,3,5-TrCBz, PeCBz, HxCBz, and PCDD/Fs, where 1,3,5-TrCBz showed the strongest relationship with toxic PCDD/Fs congeners.

To conduct a more detailed analysis of waste types, Fig. 4(b) shows the result of PCA analysis. The combined contribution of the two factors reaches 98.9 %, with Factor 1 accounting for 86.5 %. The result suggests that the two factors can represent most of the information in the original variables, and the correlations between these waste types are relatively strong. These waste-type conditions can categorized into two main groups: DW1 and DW4 conditions are considered one group, and DW2, DW3, WW, and MW conditions are considered another group. It proves that adding waste with high chlorine concentration such as waste resin, PVC, and rubber to the plasma treated waste alone will have a similar impact on the emission concentration of CBzs and PCDD/Fs in the stack gas. These will lead to increased concentrations of CBzs and PCDD/Fs emissions. The synergistic effect after the joint addition of PVC and rubber (DW4) results in its impact on CBzs and PCDD/Fs emissions being different from that of adding alone, but similar to the emission characteristics of pure miscellaneous dry waste.

The PCA results showing the relationship between CBzs, PCDD/Fs, traditional pollutants concentrations, and operational parameters under six waste types conditions are presented in Fig. 4(c), with Factor 1 contributing 41.9 %. In the PCA result, CBzs concentrations were close to PCDD/Fs I-TEQ values, with CBzs showing a stronger correlation with PCDDs than PCDFs. This finding aligns with the Pearson coefficient analysis results in this study, but differs from previous findings in MSWIs [38]. Moreover, the combustion chamber air temperature, filter inlet pressure, and CO_2 concentration were positioned far from PCDD/Fs I-TEQ values and showed negative signs in Factor 1 and 2, indicating negative correlations with PCDD/Fs emissions. In addition, the differential pressure between the quench tower and filter as well as the stack flue gas temperature were closely associated with PCDD/Fs concentrations and I-TEQ values, suggesting their potential role as influencing factors in PCDD/Fs formation.

As shown in cluster dendrogram (Fig. 5), hierarchical cluster analysis reveals the similarities and differences among six simulated nuclear power plant waste types, highlighting the relationships among these variables. These waste types could be assigned to two broad categories, which is consistent with the PCA analysis results for waste types. In more detail, DW2, DW3, WW, and MW can also be divided into two sub-categories, where DW2, DW3, and WW are one sub-category, and WW

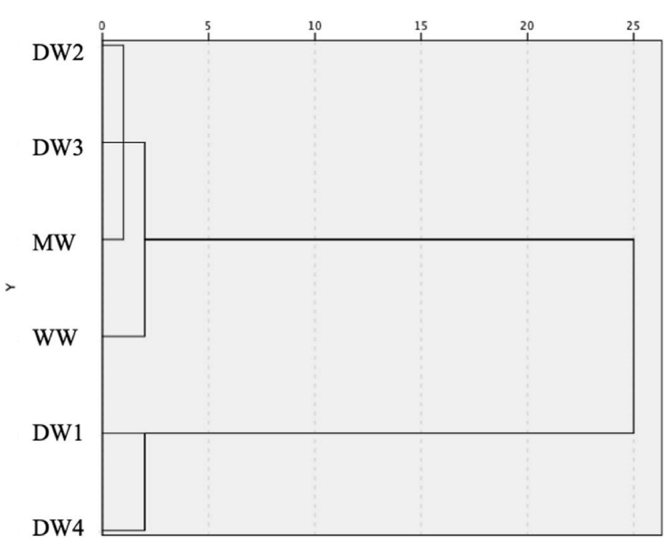

Fig. 5. Hierarchical cluster analysis of simulated nuclear power plant waste samples.

is the remaining sub-category. DW2, DW3, and MW are closely related to each other and show strong correlations with WW, which indicates that adding high chlorine content wastes such as rubber, PVC, and waste resin to ordinary miscellaneous dry wastes will have similar impacts on the emission of CBzs and PCDD/Fs in the flue gas. In addition, WW condition has only waste resin as one kind of waste, which is different from the DW2, DW3, and MW conditions with multiple waste types mixed with miscellaneous dry wastes. However, due to the existence of waste resin, there is still a great correlation between the WW condition and the DW2, DW3, and MW conditions. Meanwhile, as mentioned in the previous PCA results for waste types, the synergistic effect of PVC and rubber in DW4 condition results in its emission characteristics of CBzs and PCDD/Fs in the flue gas being similar to those of pure miscellaneous dry waste, but different from those of wastes with high chlorine content such as rubber, PVC, and waste resin. Therefore, when disposing of wastes containing high chlorine content, adding substances that can act synergistically with them is an effective means of controlling PCDD/Fs emissions.

3.3.2. *Pearson analysis*

According to the distribution characteristics of CBzs and PCDD/Fs in stack flue gas and the PCA analysis results, 1,3,5-TrCBz, PeCBz, HxCBz, the differential pressure between quench tower and filter, and stack flue gas temperature were closely associated with PCDD/Fs I-TEQ values. To clarify key factors strongly influencing the formation of PCDD/Fs under different simulated radioactive nuclear waste types, Pearson correlation analysis [39–41] was performed on CBzs, PCDD/Fs, traditional pollutants concentrations, and operating parameters. The Pearson correlation coefficients are illustrated in Fig. 6, with detailed correlation coefficient values and significance levels provided in Table S4.

The results in Fig. 6(a) indicated that CBzs exhibited stronger correlations with PCDD/Fs I-TEQ values under DW2, total DW, and total conditions than those in DW1, DW3, DW4, and MW conditions. This suggested that CBzs may have a closer relationship with PCDD/Fs I-TEQ values under comprehensive conditions with more samples. In addition, when rubber was added to the miscellaneous dry waste (DW2), the correlation between CBzs and PCDD/Fs I-TEQ values was negative, which was different from other conditions. One possible reason is that the presence of sulfur in rubber inhibits the formation of CBzs, resulting in a negative correlation between CBzs and PCDD/Fs. Another possible reason is that some components in rubber will compete to consume chlorine in waste, reducing the contribution of chlorine to PCDD/Fs, thereby reducing the amount of PCDD/Fs generated. However, this will lead to an increase in CBzs concentration, ultimately showing a negative correlation with PCDD/Fs. The study also found that high-chlorinated CBzs were more strongly associated with PCDD/Fs I-TEQ values than low-chlorinated ones under total conditions, consistent with findings from previous studies in HWIs [28] and MSWIs [38]. Among these, 1, 4-DCBz exhibited the strongest correlation with PCDD/Fs I-TEQ values ($r = 0.58$) under total conditions. Moreover, under DW2 condition in this plasma melting furnace, 1,2,3,5-TeCBz and HCBz showed a strong negative linear correlation, with $r > 0.75$.

As shown in Fig. 6(b), the correlation between traditional pollutants and PCDD/Fs I-TEQ values is generally positive under the total conditions, but this is the opposite under the DW2 condition. This is similar to the research results on the correlation between CBzs and PCDD/Fs mentioned above, both of which are due to the presence of a small amount of sulfur in rubber and the consumption of chlorine sources. In addition, HCl, CH_4, O_2, and CO showed strong correlations with PCDD/Fs I-TEQ values under specific working conditions. The concentrations of CH_4 exhibited a negative correlation with PCDD/Fs I-TEQ values under DW2 condition ($r = -0.91$), but a positive correlation under DW4 condition ($r = 0.99$). HCl also demonstrated a strong negative correlation with PCDD/Fs I-TEQ values under DW2 condition, with a correlation coefficient close to -1, which proved that the presence of rubber has a consumption effect on the chlorine sources. These findings

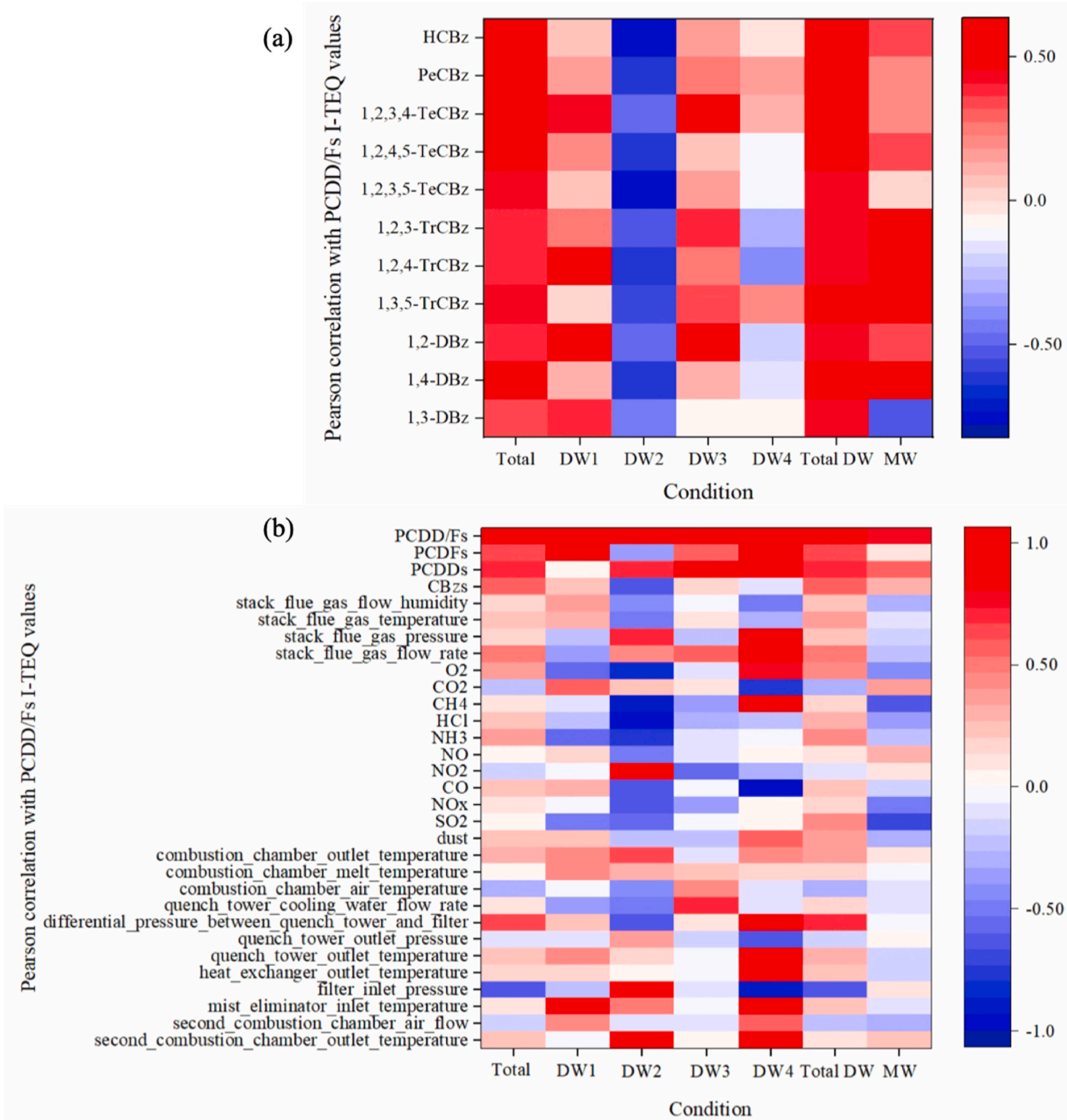

Fig. 6. Pearson correlation analysis between (a) CBzs concentrations and PCDD/Fs I-TEQ values; (b) traditional pollutants, operating parameters, and PCDD/Fs I-TEQ values.

suggested that lowing chlorine concentrations could play a crucial role in reducting PCDD/Fs emissions [42].

Meanwhile, it must be mentioned that PCDDs exhibited a stronger correlation with PCDD/Fs I-TEQ values than PCDFs under total conditions. This result revealed that PCDDs are more likely to be generated during the plasma treatment of these types of wastes, or that PCDDs are more difficult to degrade than PCDFs in this process, making their contribution to the overall I-TEQ values more significant. What cannot be ignored is that under the DW2 condition with the addition of rubber, PCDFs showed a negative correlation with PCDD/Fs I-TEQ values, indicating that the addition of rubber may be able to inhibit the formation of PCDFs, or PCDFs may be more easily degraded by pyrolysis or plasma than PCDDs.

The analysis of the correlation between operational parameters and PCDD/Fs concentrations revealed that most operational parameters exhibited weak linear correlations with PCDD/Fs I-TEQ values, except for the differential pressure between the quench tower and filter and filter inlet pressure. The Pearson coefficient for the differential pressure between the quench tower and filter and PCDD/Fs I-TEQ values was positive under total conditions ($r = 0.61$), and it reaches close to 1 under DW4 condition. The pressure difference between the quench tower outlet and the filter inlet can reflect the airflow velocity and obstruction of the flue gas through APCDs. If the pressure difference is too large, the airflow will be blocked, and particulate matter will gather or settle in the pipeline, increasing the contact opportunity between PCDD/Fs precursors and the catalytic surface, thereby promoting PCDD/Fs formation. Additionally, it prolongs the residence time of waste gas within the temperature range suitable for PCDD/Fs formation (250-500 °C), which is more conducive to the heterogeneous precursor reactions and de novo synthesis of PCDD/Fs [43].

4. Conclusion

To provide guidance on waste compatibility for low-level PCDD/Fs releases during the plasma melting disposal of radioactive waste, and to ensure the results are broadly applicable and representative, emission characteristics of thirty-two pairs of CBzs and PCDD/Fs, along with their correlations from stack gas, were analyzed from a plasma high-temperature melting furnace under different waste types conditions (rubber, PVC, and waste resin).

Adding high-chlorine materials like waste resin, PVC, or rubber to pure miscellaneous dry waste individually increased CBzs and PCDD/Fs emissions in stack gas to a maximum value of 0.408 ng TEQ/Nm^3. Among them, the addition of rubber to miscellaneous dry waste has the greatest impact on I-TEQ values, especially the increase of PCDDs. However, the simultaneous addition of PVC and rubber to the waste will produce a synergistic effect, resulting in a reduction in PCDD/Fs emissions. 2,3,7,8-TCDD (34.9 %–54.1 %) and 2,3,4,7,8-PeCDF (27.0 %–47.2 %) were the predominant contributors of PCDD/Fs I-TEQ values under all conditions. Operational analysis identified the differential pressure between the quench tower and filter as a key factor ($r = 0.61$) influencing PCDD/Fs formation, while 1,4-DCBz exhibiting the strongest linear correlation ($r = 0.58$) among all CBzs congeners. PCA and cluster analysis indicated that individually adding waste resin, PVC, or rubber to the waste treated by plasma similarly promoting CBzs and PCDD/Fs emissions, whereas the emissions of combined PVC and rubber addition were comparable to pure miscellaneous dry waste.

This study benefits guiding the safe disposal of radioactive waste and optimizing of plasma technology process during the plasma melting disposal of radioactive waste and provides effective reduction strategies. Specifically, the co-addition of PVC and rubber is effective in reducing PCDD/Fs emissions, which provides guidance on a cost-efficient strategy for the disposal of high-chlorinated radioactive waste. In terms of operational parameters, it's important to maintain optimal airflow through APCDs to prevent the build-up of catalysts, particulate matter, and other substances, considering the role of differential pressure. In the future, systematic investigations of more waste types and compatibility conditions will be conducted to optimize waste mixing strategies for better control of PCDD/Fs emissions during the disposal of radioactive waste using plasma melting technology. Meanwhile, these findings will be validated in a full-scale plasma facility treating actual radioactive waste, as well as exploring the unique effects of radiological contamination on emission profiles.

CRediT authorship contribution statement

Wenqian Jiang: Writing – original draft, Visualization, Software, Methodology, Formal analysis, Conceptualization. **Lulu Dong:** Writing – review & editing, Resources. **Minghui Tang:** Writing – review & editing, Supervision, Project administration. **Shengyong Lu:** Writing – review & editing, Supervision. **Fanjie Shang:** Resources. **Jie Lin:** Investigation. **Shaofu Tang:** Methodology.

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.joei.2025.102085.

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