



Absorption of PCDD/F Emissions in Flue Gas Using Activated Carbon Injections in South Africa

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<https://doi.org/10.18280/ijstdp.190709>

ABSTRACT

Received: 1 March 2024
Revised: 5 May 2024
Accepted: 10 June 2024
Available online: 30 July 2024

Keywords:

PCDD/Fs, activated carbon, removal efficiency, flue gas, incineration plant

South Africa is currently confronted with significant challenges related to the compliance of incineration facilities as incinerators are a major source of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and have become a significant issue in both environment and public health. The objective of this study was to assess the effectiveness of activated carbon injections in absorbing PCDD/Fs from the flue gas emissions. The Σ TEQ was initially calculated at 46.36 ng I-TEQ Nm⁻³ but saw a substantial reduction to 10.64 ng I-TEQ Nm⁻³ following AC treatment. This study was able to achieve an overall removal efficiency of 85%. Notably, highly chlorinated congeners exhibited higher removal efficiencies, indicating the selective adsorption properties of activated carbon. It was also observed that there was a correlation between PCDD/F congeners and flue gas compositions during the activated carbon treatment as congeners reacted with HCl to reduce the chlorine in synthesis of PCDD/Fs. While the activated carbon injections did lead to a notable reduction in PCDD/Fs, the surface area and porosity of the activated carbon also influenced removal efficiency. It is recommended that activated carbon with higher absorption capacity be used to enhance overall efficiency, with further modification to increase surface area and absorption for even greater effectiveness. However, despite the promise shown by activated carbon treatment in reducing PCDD/F emissions, further optimization and technological advancements are necessary to meet stringent regulatory standards.

1. INTRODUCTION

South Africa is facing a significant challenge in managing its municipal solid waste with reports showing a staggering 122 million tons is generated annually, presenting a formidable waste management issue that demands serious attention and solutions [1]. About 70% of that waste goes to landfill sites and incineration facilities [2]. Incinerators are a major source of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and have become a significant issue in both aspects of environmental and public health protection [3, 4]. There are three distinct sources that contribute to the emergence of PCDD/Fs within incineration processes. Firstly, PCDD/Fs arise as trace constituents inherent to the waste itself. A fraction of these compounds endures the incineration procedure and is subsequently conveyed within the flue gas. However, the quantitative contribution of PCDD/Fs from this origin remains relatively minor in comparison to alternative sources [5]. Secondly, the source of PCDD/F formation emanates from pyrolysis processes involving precursors present in the waste such as polychlorinated biphenyls (PCBs), chlorinated phenols, and chlorinated benzenes. These precursors undergo chemical

transformation during incineration leading to the generation of PCDD/Fs [6]. Lastly, this source involves the phenomenon of de novo synthesis. This intricate process yields PCDD/Fs from materials not intrinsically linked to these compounds. This synthesis occurs as a result of heterogeneous catalytic reactions occurring on fly ashes during the cooling of gases [7].

PCDD/Fs are highly toxic compounds that can pose significant health risks to humans and the environment upon exposure. Long-term exposure to low levels of PCDD/Fs has been associated with adverse effects on the immune system, reproductive system, and development, as well as an increased risk of cancer [8, 9]. According to Bo et al. [10], PCDD/Fs can bioaccumulate in the food chain, leading to higher concentrations in organisms at the top of the food web. This bioaccumulation can result in adverse effects on wildlife, including reproductive abnormalities, developmental deformities, and population declines. While Song et al. [11] stated that, PCDD/Fs can persist in the environment for long periods, accumulating in soil and sediment. Contamination of soil and water bodies with PCDD/Fs can impact ecosystem health and biodiversity, as well as pose risks to human health through the consumption of contaminated food and water [11]. The same can be assumed in South Africa, however, there is a

lack of comprehensive data on the dietary intake of dioxins in Africa, particularly due to limited information regarding PCDD/F emissions and food consumption quantities among different age groups in each country and the environmental impact of PCDD/Fs [12].

On May 22, 2001, the Stockholm Convention signed the proposed reduction agreement for these persistent organic pollutants [13, 14]. Among the significant outcomes of this convention, a crucial resolution emerged that mandates the elimination/ or reduction of the release of PCDD/Fs [15]. European countries took measures to address the PCDD/Fs problem earlier than African regions, with MSW incinerators as a part of countermeasures to address PCDD/Fs pollution problems [16]. However, the development of the incineration industry in Africa has lagged behind the developed countries. Recently there has been a recent strong push from developing countries to encourage the establishment of incineration facilities [17]. Hence, it is imperative to regulate the release of PCDD/Fs from hazardous and medical waste incinerators. Among the typical control technologies for PCDD/F emissions, activated carbon (AC) adsorption is the predominant control measure used to remove dioxins from flue gas [18]. Its effectiveness varies depending on factors such as the rate of carbon injection, injection technique, carbon characteristics, flue gas temperature, and particulate matter control approach [19]. Most available AC variants exhibit a substantial specific surface area and numerous micropores resulting in a wide spectrum of adsorption characteristics [20]. The efficacy of activated carbon's adsorption is heavily influenced by its pore configuration and the chemical properties of its surface area [21].

The inconsistencies in previously published studies on the impact of AC on the removal efficiency of PCDD/Fs have been noted. Various studies such as Wang et al. [21], Cerasa et al. [22], and Ren et al. [23] found that variability in AC properties played a big role in the removal efficiency. Different studies used AC with varying properties such as surface area, pore size distribution, and chemical composition. These differences significantly influence the adsorption capacity and efficiency of AC for PCDD/Fs removal [21]. Studies employing AC with different characteristics could yield conflicting results due to these inherent variations [5, 23]. While other inconsistencies were noticed, Ma et al. [24], and Li et al. [25] stated that differences in experimental conditions such as temperature, gas flow rate, residence time, and PCDD/Fs concentration in the flue gas, can impact the performance of AC in removing PCDD/Fs. Studies conducted under disparate conditions yield conflicting findings due to the varying degrees of PCDD/Fs adsorption onto AC [26]. According to Hsu et al. [27] and Wei et al. [28] flue gas from waste incineration contains a complex mixture of pollutants, including PCBs, PAHs, heavy metals, and particulate matter. The presence of these interfering compounds can compete with PCDD/Fs for adsorption sites on AC, thereby influencing the overall removal efficiency [28]. Studies that do not account for these interfering compounds report inaccurate or inconsistent results regarding the effectiveness of AC for PCDD/Fs removal [5, 23, 25]. The design and operation of air pollution control devices, including the placement and configuration of AC injection points, as well as the integration of other control technologies such as bag filters and scrubbers, impact the performance of AC for PCDD/Fs removal [25, 29].

Apart from the adsorption of PCDD/Fs from gas streams, AC can paradoxically contribute to the formation of PCDD/Fs

through de novo synthesis. This unintended consequence can exacerbate the challenge of disposing of spent sorbent material [30]. Carbon adsorption is generally more efficient at reducing lower chlorinated dioxins with higher gaseous fractions than highly chlorinated ones [31]. This involves injecting the amount of solid adsorbent into the flowing gas, providing sufficient contact time for the efficient use of the carbon, and then separating the adsorbent before discharging the gas up the stack [31]. Nonetheless, a comprehensive investigation into the removal efficiencies of PCDD/Fs and chlorobenzenes (CBzs) utilizing various technologies remains lacking. The efficacy of the semi-dry flue gas desulfurization (SDFGD) system, AC injection, bag filters (BF), the selective catalytic reduction (SCR) reactor, and the wet scrubber in eliminating PCDD/Fs has not been thoroughly investigated [6]. Previously published studies concerning the impact of AC on the removal efficiency of PCDD/Fs present inconsistencies in their findings as discussed above. South Africa is currently confronted with significant challenges related to the compliance of air quality from incineration facilities. The objective of this study was to assess the effectiveness of AC injections in absorbing PCDD/Fs from the flue gas emissions of a medical waste incinerator. The data obtained from this study will proactively address the compliance issues and contribute to the sustainability and continued operation of incineration facilities.

The current study presents groundbreaking findings aimed at addressing critical environmental and public health concerns. By investigating the effectiveness of AC injections in reducing PCDD/Fs emissions from a medical waste incinerator in South Africa, the study directly confronts the significant risks posed by PCDD/Fs compounds. The study was of paramount importance due to the urgent need for incineration facilities to comply with regulatory standards regarding PCDD/F emissions, thereby ensuring environmental protection and public health safety. Moreover, the study's innovative approach to pollution control through AC injections contributes to the development of sustainable and cost-effective strategies for reducing PCDD/F emissions from waste incinerators. By providing empirical data on the performance of AC injections, the current study advances scientific knowledge in the field of pollution control and informs future efforts to improve the efficiency and effectiveness of pollution control technologies in incineration processes. Moreover, this paper represents a significant step forward in addressing PCDD/F emissions and promoting environmental sustainability in South Africa's waste management practices.

2. METHODS

2.1 Plant description

The study was conducted in medical waste incineration facilities. The site is positioned in a traditional industrial and commercial zone area with a residential area a kilometre away. In this facility, the combustion of medical waste happens in two phases. In the first phase, waste is sustained into the primary chamber where gases are produced during combustion (Figure 1). The second phase takes place in the secondary chamber where more temperature is applied up to 1200°C to combust gases generated in the primary chamber. This facility incinerates roughly 700 tons of medical waste every month, all originating from private and public healthcare facilities around

the country. There are four auxiliary burners to ensure the incinerator reaches the recommended temperatures, two situated at the entrance to the secondary chamber and two other burners in the primary chamber. The air pollution control devices included AC, a BF and a dry scrubber (DS) for controlling pollutant emissions. The AC was injected into the exhaust stream to remove PCDD/Fs; subsequently, the BF was placed downstream and used to remove AC particles and residual dust. This process consumed a large amount of AC thus increasing the cost of the incineration plant. The silo was filled with 50 kilograms of AC every day to extend the duration the plant stays in gravimetric mode. During sampling, two types of waste were fed into the furnace through the mechanical pump. The solid waste 700 kg was pumped into the primary chamber for nearly 2 hours and the temperature was maintained between 850 and 980°C. The calorific values for solid waste and liquid waste were 6600 kcal kg⁻¹ and 6400 kcal kg⁻¹ and the chlorine contents were 4.23% and 17%, respectively. Moreover, solid waste often has a complex composition and fluctuating calorific value so it is deemed unsuitable during startup. The temperature of the rotary kiln and secondary combustion chamber decreased sharply after feeding liquid waste which recovered loosely with the help of auxiliary burners. The variable speed drive was used to dose AC into the system and two manual blowers were used to ensure continuous flow of AC into the filtration system.

fed into the primary chamber where combustion takes place. The gases produced in the primary chamber then move to the secondary chamber for further combustion at higher temperatures.

2.2 AC properties and dosage rates

This study encompassed commercially available powdered activated carbon purchased from Lhoist South Africa PTY Ltd. To examine the influence of the physical and chemical attributes of AC on the management of PCDD/Fs, the evaluation of surface area and pore structure was executed through nitrogen adsorption at a temperature of 200°C. This process was conducted using a TriStar 3000 gas adsorption analyzer. The analytical system functioned by quantifying the extent of nitrogen adsorbed onto or desorbed from a solid sample at diverse equilibrium vapor pressures. The assessment employed the principles of physical adsorption and capillary condensation to acquire insights into the surface area and porosity characteristics (Table 1). AC injection rates were calculated and assessed to evaluate the dosage rate at which AC was most effective in reducing PCDD/Fs. Two different adsorbent dosages were applied for the investigation of optimum dose, at 1.2 kg h⁻¹ and 2.5 kg h⁻¹. The dosage rates were collected from the feeder and compared with the PCDD/Fs results to determine the rate at which the sorbent was mostly efficient. The maximum removal efficiency of 85% was achieved at 2.5 kg h⁻¹ and no further improvement was achieved at a dosage rate higher than 2.5 kg h⁻¹. Thus, there's a need to explore advanced technology to improve the removal efficiency of flue gas contaminants.

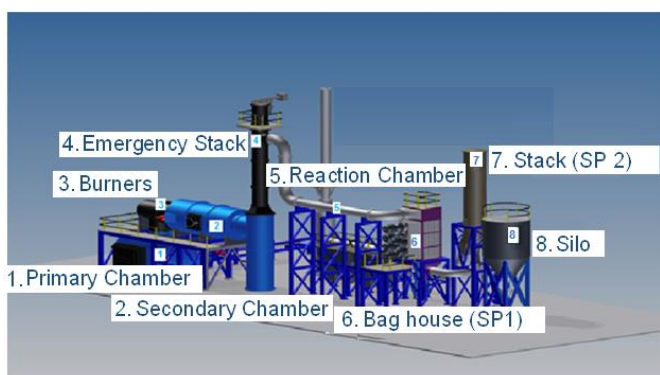


Figure 1. Schematic diagram of RD 700 medical waste incineration facility

Figure 2 represents the process flow in a medical waste incineration facility as described. The solid and liquid waste is

Table 1. Properties of AC

| Properties of AC | |
|--|-------------|
| Ash (%) | 7 |
| Moisture (%) | 8 |
| Chloride (%) | 0.2 |
| Carbon (%) | 87.5 |
| Total sulfur content (%) | 0.4 |
| Surface area (m ² g ⁻¹) | 220 |
| Pore volume (cm ³ g ⁻¹) | 0.21 |
| Micropore, pore diameter <2 nm (cm ³ g ⁻¹) | 0.04 |
| Mesopore, pore diameter 2 - 50 nm (cm ³ g ⁻¹) | 0.13 |
| Average pore diameter (nm) | 2.25 |
| Injection rate (kg h ⁻¹) | 1.2 and 2.5 |

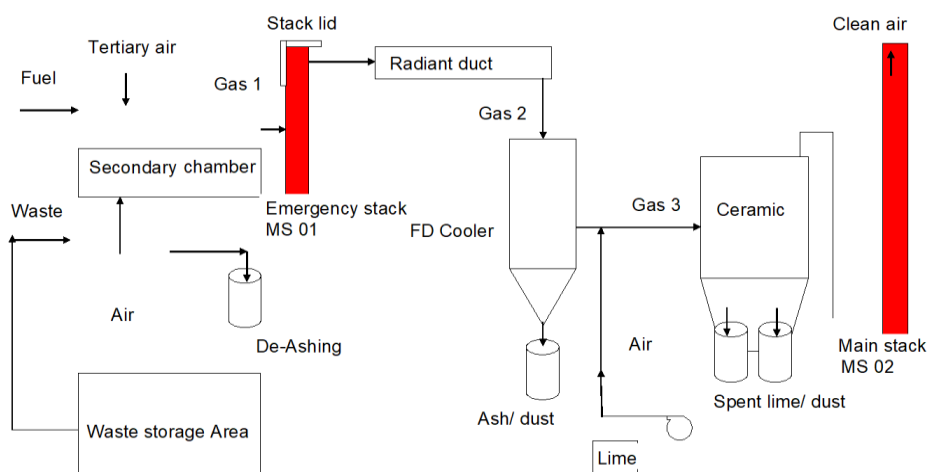


Figure 2. Incineration process flow

2.3 Sample collection, preparation and analysis

Flue gas sampling was conducted using US EPA Method 23. The gas phase sample was collected by absorption on XAD-2 resin, whereas the particle-bound sample was collected by a fiberglass filter and by rinsing the sampling probe thereafter. At a sampling rate of 0.014 m³/min (0.5 cfm) and the sample volume per 3 hours of 0.84 m³/h, the sample time was 6.25 h per sample. The XAD-2 resins were spiked with known amounts of US EPA Method 23 internal standard solution before sampling to check the PCDD/Fs sampling efficiency. The procedure for gas sampling consisted of the following steps: XAD-2 resins were placed at SP2 for a duration of 6.25 hours without AC injection. Subsequently, once the allotted time had passed, the XAD-2 resins were extracted, AC was introduced and fresh XAD-2 resin was then positioned at SP2 for another 6.25 hours. After sampling, filters and XAD-2 resin were stored and maintained in the dark until transferred to the laboratory. For a better understanding of PCDD/F distributions between the gas and particulate phases, the XAD-2 and fiber filter samples were separately Soxhlet extracted with toluene for 24 h. While the other flue gas samples were extracted with toluene together. After extraction, the toluene extract was concentrated to nearly 1 mL by rotary evaporator and replaced by 5 mL hexane. Then, the hexane solvent was treated with concentrated sulphuric acid. After being subjected to a series of clean-up columns including a hybrid silica gel column (sulphuric acid and sodium hydroxide silica gel) and an aluminum oxide column, the eluate was then concentrated to 1 mL through rotary evaporation and transferred to a vial. The solvent was further concentrated to near dryness with a pressure blowing concentrator. A known amount of Method 23 recovery standard solution was added to the solvent. The extracts were analyzed with high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS). The HRGC/HRMS was equipped with a DB-5MS fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 µm), used for separating the PCDD/F congeners. Helium was used as carrier gas. The oven temperature was programmed with an initial temperature of 150°C, increased to 280°C and then held for 20 min. The mass spectrometer was operated with a minimum resolution of 10,000 under positive EI conditions, and data were obtained in the selected ion monitoring (SIM) mode. The electron energy and source temperature were specified at 38 eV and 280°C, respectively.

2.4 Chemical standards and organic solvents

US-EPA method 1613 was used for the analysis of PCDD and PCDF. A set of five calibration standard solutions (CS1-CS5) containing the 17 toxic congeners of native PCDD/F (2,3,7,8-positional substitutes) at concentrations between 0.5 and 2000 ng/ml and corresponding ¹³C₁₂ isotope enriched PCDD/F at 100 ng/ml (except for ¹³C₁₂-OCDD which was 200 ng/ml) with a purity of P99% was used. The ¹³C₁₂ labeled standard stock solution containing 17 toxic congeners of PCDD/F at the concentration level of 100 ng/ml (except for ¹³C₁₂-OCDD which was 200 ng/ml) was used to quantify the corresponding native compounds by the isotope dilution method. This solution was added to the samples just before the extraction process. However, the XAD-II resin which was applied for trapping PCDD/F from the exhaust gas of the reactor was spiked prior to experimentation. Furthermore, an

internal standard solution containing isotopically labeled ¹³C₁₂-1,2,3,4-TCDD and ¹³C₁₂-1,2,3,7,8,9-HxCDD at a concentration level of 200 ng/ml in nonane was added to the purified extract prior to GC injection as standards for the determination of recovery percentages of labeled PCDD/Fs. All these standards were purchased from Promolab Pty Ltd T/A Separations, South Africa. All the organic solvents such as acetone, toluene, dichloromethane, n-hexane, and ethyl acetate used for extraction, clean-up and glassware rinsing, were certified grade for dioxin analysis and purchased from Lasec SA PTY Ltd.

2.5 Quality assurance

Comprehensive quality control measures were implemented throughout the PCDD/Fs analysis process to ensure the accuracy and reliability of the obtained data. Field blanks and laboratory blanks, analyzed alongside flue samples, consistently showed PCDD/Fs levels at least three times lower than the detection limit, affirming the effectiveness of sampling and analytical procedures in preventing contamination. Further credibility checks included the addition of internal standards mixture before Soxhlet extraction, allowing for the monitoring and correction of any analytical variations. Internal standard recovery rates, ranging between 85% and 130%, surpassed the required standard of 70% to 130%. Moreover, the careful monitoring of PCDD/PCDF surrogate recovery rates, maintained between 95% and 130%, aligned with the specified criteria of 70% to 130%, ensuring consistent behavior of surrogate compounds for reliable quantification. Calibration curves were regularly verified to uphold the precision of the analytical instrument collectively contributing to the overall reliability and credibility of the PCDD/Fs analysis results for environmental assessments and regulatory compliance.

3. RESULTS AND DISCUSSION

In this sampling campaign, two distinct sampling locations were used: Particulate samples were collected in the dual bag filter before and after the injections referred to as SP1 and gas phase samples were collected at the stack both before and after the AC injections, denoted as SP2. Concentrations measured before the intervention are referred to as initial concentrations (IC). The results are presented in Figures 3-6 as ng WHO₂₀₀₅ TEQ m⁻³.

3.1 PCDD/Fs congener profile and concentrations in particulate phase

At SP1, the particulate phase showed a significant presence of 1,2,3,4,7,8,9-HpCDF as the major congener with the IC of 39.84 ng/Nm³ as indicated in Figure 3. Following the introduction of AC at 1.2 kg/h injection rate, a notable reduction of 5.95 ng/Nm³ was observed. However, when the injection rate was further increased to 2.5 kg/h only minor improvement of 5.17 ng/Nm³ was reported. Congener 2,3,4,7,8-PeCDF also exhibited a high IC of 32.17 ng/Nm³. After the AC injection, the concentration decreased to 9.65 ng/Nm³ and with the subsequent 2.5 kg/h injection rate, it further reduced to 8.38 ng/Nm³. The findings indicate that at SP1, increasing the injection rate to 2.5 kg/h resulted in minimal reactions with no substantial improvement observed

in congeners. Observations indicated that the concentrations of PCDF congeners were notably higher in comparison to those of PCDD congeners. PCDF congeners and PCDD congeners have different chemical structures, with PCDFs typically being more volatile than PCDDs. The increased volatility of PCDFs may have contributed to their presence in the particulate phase making them less susceptible to capture

by the AC. This could be attributed to the substantial chlorine content present in the waste. The chlorine content in the waste played a significant role in shaping the congener profiles and the mechanisms involved in the formation of PCDD/Fs in the bag filter. When the chlorine content was elevated, PCDD/Fs tended to form through de novo synthesis with a higher prevalence of PCDFs over PCDDs [32].

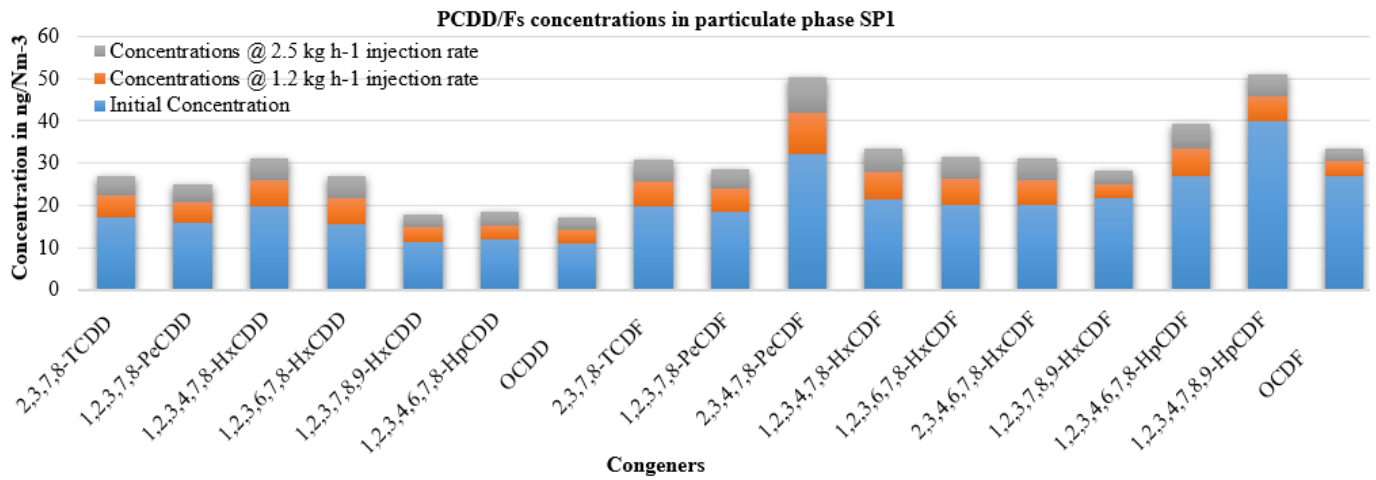


Figure 3. PCDD/Fs concentrations in particulate phase in SP1

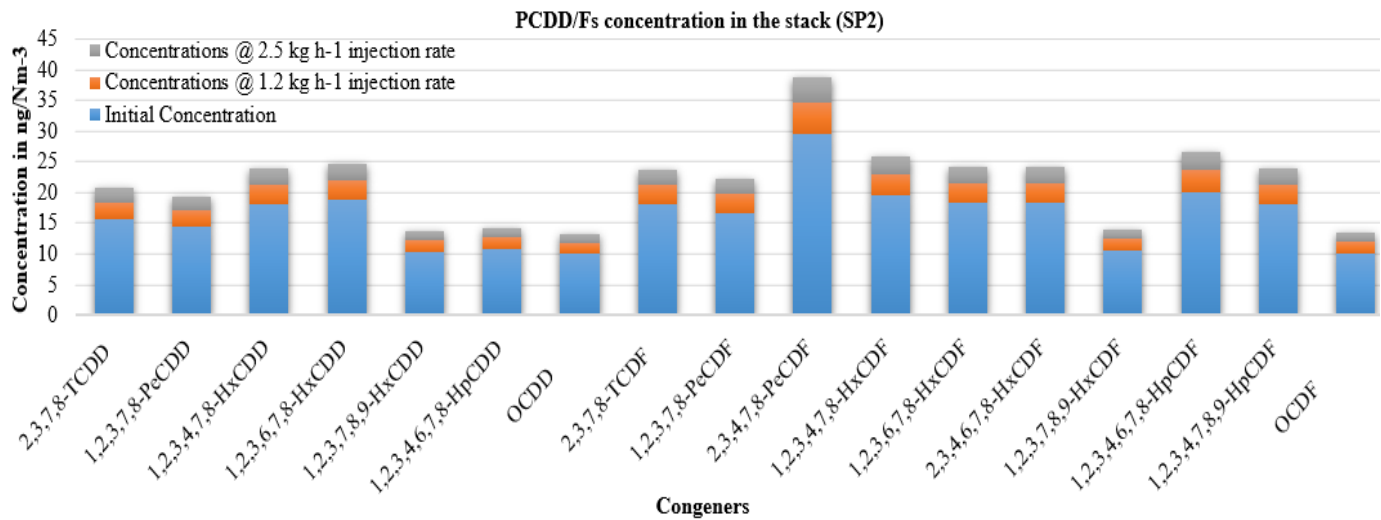


Figure 4. PCDD/Fs concentrations in gas phase in stack

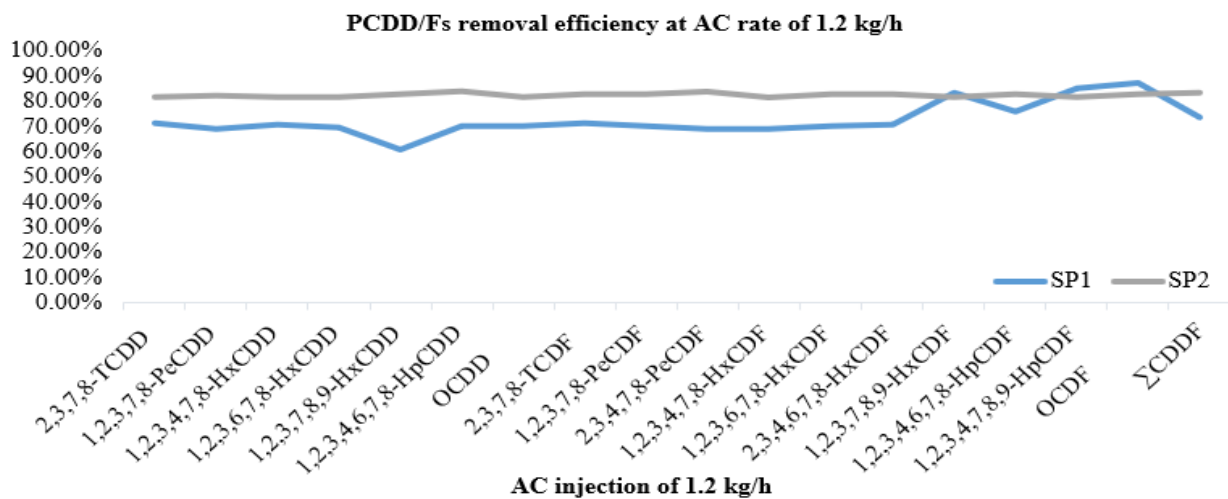


Figure 5. PCDD/Fs removal efficiency at 1.2 kg/h AC injection rate

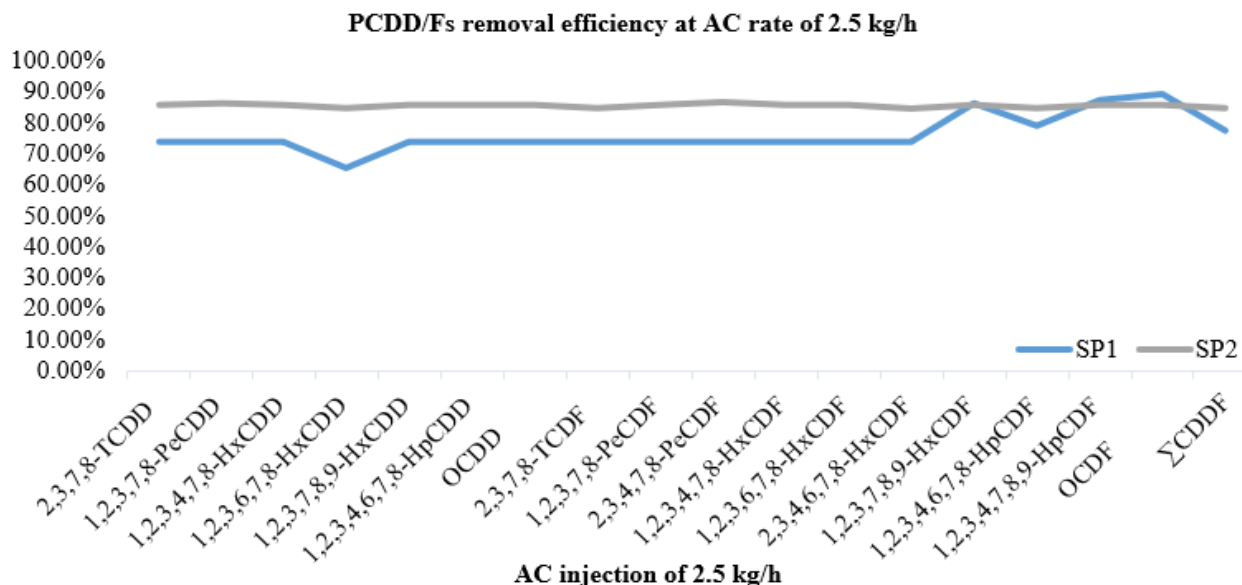


Figure 6. PCDD/Fs removal efficiency at 2.5 kg/h AC injection rate

OCDD was the least dominant congener reporting the lowest IC of 10.95 ng/Nm³. The concentration further decreased to 3.28 ng/Nm³ after the AC injection. A significant reduction of 2.85 ng/Nm³ was observed when the injection rate was increased to 2.5 kg/h. In contrast to the results reported by Fu et al. [33] where 2,3,7,8-TCDD was identified as the least dominant congener with concentrations ranging between 0.17 and 0.02 ng/Nm³. Similar trends in reduction were observed in several congeners including 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,7,8-PeCDD which reported concentrations of 2.97 ng/Nm³, 3.08 ng/Nm³ and 4.15 ng/Nm³ respectively. It was observed that the highly chlorinated congeners were detected higher than that of low chlorinated congeners in the particulate phase. Lower chlorinated congeners in contrast to their higher chlorinated counterparts exhibit greater vapor pressures. Consequently, they have a propensity to transition into the gas phase thereby increasing the likelihood of adsorption onto AC. AC displayed varying degrees of adsorption selectivity among PCDD/F congeners exhibiting different preferences for adsorption whereas other congeners showed limited or inadequate selectivity for AC adsorption. When comparing the PCDD/Fs emissions in SP1, it was evident that the concentration had significantly reduced by almost an order of magnitude from ΣCDDF 350.17 ng/Nm³ to 79.28 ng/Nm³. While the control measures had a noticeable effect, it fell short of meeting the stringent requirements. This confirms that the combination of AC injection and a single bag filter system was insufficient to reduce PCDD/Fs emissions to the desired level of 0.01 ng/Nm³.

The findings from the current study indicate a substantial reduction in PCDD/Fs concentrations in the particulate phase following the introduction of AC injections. This reduction is particularly notable for congeners such as 1,2,3,4,7,8,9-HpCDF and 2,3,4,7,8-PeCDF, which exhibited significant reduction in concentration after AC injection. The observed reduction in PCDD/Fs concentrations highlights the effectiveness of AC injections in mitigating PCDD/F emissions from waste incineration processes. Furthermore, the data reveal insights into the complex interplay between PCDD/F congeners, waste composition, and AC adsorption selectivity. The higher concentrations of PCDF congeners compared to PCDD congeners suggest the influence of waste

chlorine content on congener profiles and de novo synthesis mechanisms. Additionally, the varying degrees of adsorption selectivity among PCDD/F congeners highlight the importance of considering congener-specific properties when designing pollution control strategies. Despite the significant reduction in PCDD/F concentrations, the data also indicate that the combination of AC injection and a single bag filter system may not be sufficient to meet stringent regulatory requirements. This highlights the need for further optimization and potentially the integration of additional control measures to achieve desired emission levels. Overall, the findings provide valuable insights into the effectiveness and limitations of AC injection technology in mitigating PCDD/F emissions, informing future research and pollution control strategies in waste incineration facilities.

3.2 PCDD/Fs congener profile and concentrations in gas phase

In SP2, 2,3,4,7,8-PeCDF emerged as a dominant congener with the IC of 29.42 ng/Nm³ which decreased to 5.15 ng/Nm³ after the AC injection. When the injection rate was raised to 2.5 kg/h, the concentration experienced a slight reduction to 4.21 ng/Nm³ as indicated in Figure 4. It was noted that in the stack emissions both PCDD and PCDF congeners including 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,7,8,9-HpCDF all exhibited a similar trend. Similar congener profiles were reported for municipal and industrial incinerators by Gunes et al. [34], Bian et al. [35]. This trend was attributed to higher chlorinated PCDDs having a lower affinity for the AC surface leading to their dominance in the treated effluent. This observation is in contrast to the findings reported by Wang et al. [21] that lower chlorinated PCDD/Fs and PCDFs exhibited superior removal efficiencies in comparison to their highly chlorinated counterparts. This phenomenon can be credited to the heightened competitive nature of highly volatile congeners in AC adsorption when compared to congeners with lower volatility.

OCDD continued to be the least dominant congener with the IC of 9.98 ng/Nm³. Following the AC injection, a reduction of

1.75 ng/Nm³ was observed and this reduction further increased to 1.43 ng/Nm³ when the injection rate increased to 2.5 kg/h. This indicates that the PCDD/Fs formation was dominated by de novo synthesis and the chlorobenzene route synthesis. Highly chlorinated congener such as OCDD has a lower vapor pressure as compared to a lower chlorinated congener and has a higher tendency to condense on gas phase. Other factors like temperature and contact time with the AC played a role in the reduction. It was also observed that the removal efficiency of high chlorinated congeners increased more in the stack compared to lower chlorine content congeners. This was attributed to the preferential adsorption of lower chlorinated PCDD/F congeners as they tend to have higher vapor pressures and are less readily adsorbed. The Σ CDDF also reduced from 277.40 ng/Nm³ to 39.81 ng/Nm³ after the AC injections. The Σ TEQ was initially calculated at 46.36 ng I-TEQ Nm⁻³ but saw a substantial reduction to 10.64 ng I-TEQ Nm⁻³ following AC treatment. This was as a result that the rising vapor pressure led to the increased release of PCDD/Fs into the vapor phase and the high amount of chlorine content in the waste and the de novo synthesis in the incineration. In SP2, the IC was notably lower than what was observed in the first bag filter (SP1). It was observed that the distribution of PCDD/Fs underwent minimal reduction change as the flue gas passed through the dual bag filter, in comparison to the composition observed in SP1. This slight alteration was attributed to the filtration system in SP1, resulting in a modest IC reduction in SP2. It was concluded that while substantial improvement was evident following the AC injection treatment in SP2, the dual filter system also played a contributing role.

Based on the findings it was concluded that PCDD/F compounds were formed and controlled by many factors including the chlorine content of combusted waste, the type of rotary kiln, the operating conditions, the type and location of air pollution control equipment. Highly chlorinated congeners had relatively higher vapor pressures making them more prone to adsorption on the AC surface. They are generally more stable and less prone to transformation reactions during AC treatment ensuring their effective removal [30]. In accordance with the results obtained from the current study, AC exhibited selective adsorption properties favoring the removal of highly chlorinated congeners over less chlorinated ones. This selectivity contributed to the reduction of 1,2,3,6,7,8-HxCDF; 2,3,4,6,7,8-HxCDF; 1,2,3,7,8,9-HxCDF and OCDF concentrations which were initially had higher concentration. Both gas and particulate AC treatments effectively reduced the concentrations of various congeners, with OCDD being the least dominant congener in both particulate and gas phases. The reduction in concentration was generally more pronounced in the gas phase suggesting higher effectiveness at the higher temperature for these specific congeners. Nevertheless, further reduction is still necessary to achieve the minimum compliance threshold of 0.01 ng/Nm³ established in SA.

3.3 PCDD/Fs removal efficiencies at different AC injection rates

The removal efficiency results are presented in Figure 5 and Figure 6. At the AC injection rate of 1.2 kg/h, the overall removal efficiency at SP1 was 73%. Notably, OCDF achieved the highest reduction of 87%, followed closely by 1,2,3,7,8,9-HxCDF with 83% reduction, while 1,2,3,6,7,8-HxCDD had the lowest reduction of 60%. In the stack (SP2), an impressive

overall removal efficiency of 82% was observed and all congeners exhibited a consistent trend in their reductions, similar to results were reported by Vunain et al. [36]. Based on these findings it was concluded that AC treatment was successful in removing a significant portion of PCDD/F compounds from the initial samples. The adsorption of AC was influenced by the surface area and by the distribution of micropores and mesopores that resulted in less absorption of other compounds. As a result, properties related to the physical and chemical characteristics of congeners such as their size and molecular weight, played a role in determining the partitioning and removal mechanisms. The highly chlorinated PCDF congener exhibited a substantial decrease in concentration after AC treatment. The reduction was attributed to the efficient adsorption of highly chlorinated congeners by AC. At the AC injection rate of 2.5 kg/h, the overall removal efficiency at SP1 increased to 77%, marking a more than 5% improvement compared to what was achieved at 1.2 kg/h injection rate Figure 6. Remarkably, OCDF achieved the highest removal efficiency of 89%, while 1,2,3,6,7,8-HxCDD exhibited the lowest efficiency at 65%. In SP2, a consistent removal efficiency of 85% was achieved marking a more than 3% improvement compared to what was achieved at 1.2 kg/h injection rate.

When comparing the performance of the AC injection rate of 2.5 kg/h to 1.2 kg/h injection rates across all congeners, there was a notable improvement in the removal efficiency. OCDF consistently exhibited the highest removal efficiency, while 1,2,3,6,7,8-HxCDD consistently had the lowest removal efficiency. The higher injection rate of 2.5 kg/h resulted in enhanced removal efficiency for all congeners, demonstrating the effectiveness of this injection rate in reducing the presence of PCDD/Fs compounds in the stack gas. It was observed that the removal efficiency for PCDD/F compounds was generally higher in the gaseous phase compared to the particulate phase. The difference can be attributed to the distinct behaviors of the PCDD/Fs compounds in these two phases. It appeared that PCDD/F congeners were more effectively destructed and degraded in the gaseous phase while they tended to be absorbed and trapped by particulate phase. In the gas phase, less chlorinated PCDD/F compounds undergo processes such as thermal decomposition, reaction with reactive species or photo-degradation which can lead to their degradation and subsequent reduction in concentration. This behavior is especially pronounced when oxygen is present [21]. On the other hand, in the particulate phase, PCDD/F compounds can become physically adsorbed or captured by solid particulate matter such as fly ash or AC residues. The surfaces of these particles provide ample opportunities for adsorption and the PCDD/F compounds can attach to them effectively reducing their presence in the gas stream. This difference in behavior between the gaseous and particulate phases highlights the necessity for additional study to comprehensively investigate the presence of PCDD/Fs in ash and AC residue during the AC treatment.

The Σ TEQ was initially reported at 46.36 ng I-TEQ Nm⁻³ but was significantly reduced to 10.64 ng I-TEQ Nm⁻³ after the AC injections. These results are 2-fold higher than the observations made by Gunes et al. [34] in a study conducted in 50 different incinerators in Korea. But similar to the Σ TEQ value reported by Açıkıldız et al. [37]. The findings in this study suggest that, PCDF congeners responded better to AC treatment compared to PCDD congeners with the highest removal efficiencies achieved in gas phase. The 85% overall

removal efficiency attained in this study did not achieve the removal efficiency reported by Chai et al. [38] who found an overall efficiency of 92% and a reduction in toxic equivalent of 48%. The differences in removal efficiency among congeners may be attributed to variations in their physicochemical properties, including molecular weight, vapor pressure and adsorption affinity. These properties affected the congeners behavior during AC treatment. AC exhibited selective adsorption properties, favoring the removal of certain congeners over others. The selectivity was influenced by factors such as congener chlorination levels and molecular properties. The removal efficiencies of PCDD/F congeners in the stack were nearly similar in both injection rates. The high removal efficiency relied on gas-solid phase proportion of PCDD/F congeners and the adsorption capacity of AC to PCDD/F congeners. The surface area and porosity of the AC also affected removal efficiency. While the 2.5 kg/h AC injections did lead to a notable reduction in PCDD/Fs, it is strongly advised to utilize activated carbon with a higher absorption capacity to enhance overall efficiency. For a higher efficiency, it is suggested that AC be modified to increase the surface area and absorption. AC with a high surface area will provide more binding sites for adsorption, which can enhance removal efficiency.

During the current study, inconsistencies were observed in the variation in the effectiveness of AC in reducing PCDD/F concentrations between different congeners and phases (particulate vs. gas). While AC treatment led to significant reductions in certain congeners, such as 1,2,3,4,7,8,9-HpCDF in the particulate phase and 2,3,4,7,8-PeCDF in the gas phase, other congeners showed only minor improvements or no substantial reduction despite increasing the AC injection rates. These inconsistencies suggest that the efficacy of AC in capturing PCDD/F congeners may be influenced by factors such as their chemical structure, volatility, and affinity for adsorption onto AC surfaces. Additionally, the inconsistency in the reduction of PCDD/F concentrations between particulate and gas phases highlights the complexity of the removal process and the need for further optimization. While certain congeners exhibited greater reductions in the gas phase, indicating higher effectiveness at higher temperatures, others showed more pronounced reductions in the particulate phase. This highlights the importance of considering the specific characteristics of each congener and phase when evaluating the performance of AC treatment. Furthermore, the observed variations in PCDD/F concentrations in this study are consistent with the findings by Wang et al. [21], Cerasa et al. [22], Ren et al. [23], Ma et al. [24], Li et al. [25] confirming inconsistencies in the overall impact of AC treatment on reducing PCDD/F emissions. The current study concludes that, these inconsistencies may arise due to differences in experimental conditions, waste composition, incineration processes, and the types of air pollution control equipment used.

In conclusion, the study indicates the distribution and changes of individual PCDD/F isomers were identified before and after AC injection. For instance, at SP1, significant concentrations of specific isomers like 1,2,3,4,7,8,9-HpCDF and 2,3,4,7,8-PeCDF were observed initially, with notable reductions following AC injection. However, the reductions varied among different isomers and were influenced by factors such as injection rate and chemical properties of the isomers.

Isomers with higher chlorination levels generally exhibited lower reduction rates compared to less chlorinated ones. Moreover, the distribution of isomers differed between particulate and gas phases, highlighting the importance of considering phase-specific behavior in PCDD/F analysis. During the current study, several advantages of AC injection technology were identified. Firstly, the study demonstrated the effectiveness of AC injection in reducing PCDD/F emissions from waste incineration processes, as evidenced by significant removal efficiencies observed, reaching up to 85%. Secondly, the technology was found to offer a relatively straightforward and cost-effective approach to controlling PCDD/F emissions compared to alternative methods. Lastly, the study showed that AC injection can be implemented without significant modifications to existing incineration facilities, thereby minimizing operational disruptions. Disadvantages were also observed during the study. Firstly, AC injection required careful optimization to achieve desired removal efficiencies, as variations in removal rates among different congeners and phases were observed. Secondly, the technology necessitated proper handling and disposal of spent AC to prevent environmental contamination, posing logistical challenges. Lastly, depending on the properties of the AC used, AC injection alone may not suffice to meet stringent regulatory requirements for PCDD/F emissions, necessitating complementary control measures. On the AC scope of application, it was found that AC injection technology holds promise for application in various waste incineration facilities, including municipal and industrial settings, where PCDD/F emissions are a concern. The technology can be tailored to specific incineration conditions and emission targets through adjustments in injection rates and process parameters. AC injection complement other air pollution control measures, such as electrostatic precipitators and bag filters, to achieve comprehensive emission reduction strategies.

3.4 Correlation of PCDD/F congeners and flue gas compositions

During the study, it was noted that PCDD/Fs emissions exhibited a correlation with other gases present in the flue gas. PCDD/F compounds are typically generated through precursor and de novo synthesis reactions that occur within the post-combustion zone [39, 40]. The rates at which they are formed depend on various factors, including the levels of O₂, CO, SO₂, chlorine content in the flue gas, the temperature distribution, the presence of metals and the carbon content of fly ash [20]. During SP1, 86% of the PCDD/Fs concentration was on particulate phase after the AC injection. This high contribution from particle phase could be explained by de novo synthesis reactions and binding of high molecular weight congeners to particles. PCDD/F congeners gas/ particle partitioning were strongly influenced by vapor pressure (18.6 - 20.4%), which was proportional to temperature. Due to decreasing vapor pressure, congeners were adsorbed onto particle surfaces, especially in low temperature areas outside the bag filters. PCDD/F compounds that were not destructed within the secondary combustion chamber and were carried by gas stream towards the filtration system and adsorb onto particles in between boiler and AC reactor at temperatures between 300 and 120°C (post combustion). Operational conditions and flue gas compositions during sampling are presented in Table 2.

Table 2. Operational conditions and flue gas compositions during sampling

| Parameter | Range |
|--------------------------------------|-----------------|
| Waste loading (kg/d) | 12 500 - 16 500 |
| Primary chamber temp (°C) | 850 - 950 |
| Secondary chamber temp (°C) | 950 - 1200 |
| Flue gas composition | |
| Flow rate (dry, /Nm ³ /h) | 20 200 - 22 900 |
| O ₂ (%) | 10.22 - 11.05 |
| CO ng/Nm ³ | 9.86 - 15.10 |
| Moisture (%) | 15.6 - 20.4 |
| HCl ng/Nm ³ | 0.09 - 0.32 |
| HF ng/Nm ³ | 0.25 - 0.37 |
| NO _x ng/Nm ³ | 96 - 150 |
| NO ₂ ng/Nm ³ | 78 - 165 |
| SO ₂ ng/Nm ³ | 20.70 - 30.45 |
| Org. C ng/Nm ³ | 0.01 - 0.13 |
| PM ng/Nm ³ | 18.8 - 25.10 |
| Resident time (s) | 2 |

As the temperature dropped, the surface oxygen grouped with organic matter on the decomposed AC to release CO₂ at lower temperatures and to release predominantly CO. The CO₂ arises primarily from the decomposition of the carboxyl and lactone groups, whereas the CO results from combustion of organic matter with low oxygen content. PCDD/Fs increased again in the exit gas after AC filter as the HCl concentrations increased [14]. This increase was due to the increase of high-chlorinated PCDD/Fs in the particulate phase, whereas gas-phase PCDD/Fs showed a moderate decrease through AC. The increase in the particle bound congener concentrations was attributed to the AC content used. When the flue gas temperature in the primary chamber was 900°C, the concentration of PCDD/Fs in flue gas was 109.5 ng/Nm³. But when the flue gas temperature on the outlet on the secondary chamber decreased 850°C, the PCDD/Fs concentration in flue gas was reduced 10.89 ng/Nm³. The residence time also played a significant role in influencing the formation of PCDD/Fs. Additionally, the O₂ levels in the system had a pronounced effect on the formation of PCDD/Fs. Reducing the O₂ content hindered the advancement of the de novo reaction, resulting in decreased concentrations of PCDD/Fs. Conversely, elevating the O₂ content improved combustion efficiency and reduced the presence of organic matter during incomplete combustion.

AC treatment achieved 85% PCDD/F reduction. NO_x and NO₂ emissions also decreased by nearly 35% while particulate matter increased by 15% due to the use of AC, similar to the results reported by Themba et al. [41] when hydrated lime was used. The SO₂, NO_x, and PCDD/Fs concentrations exhibited a consistent decrease as they passed through the dual bag filter system. In contrast, the O₂ concentration showed an opposing trend, increasing along this path. Nonetheless, both SO₂ and PCDD/Fs concentrations exhibited a predominant decreasing

pattern as they traversed the APCDs. The result showed that the higher the temperature in flue gas, the less conducive to the formation of PCDD/Fs was and reducing the O₂ content in flue gas appropriately reduced PCDD/Fs emission without changing the quality of the operational conditions. The results also showed that the congeners reacted with HCl to reduce the chlorine in the synthesis of PCDD/Fs. Although the sulphurous inhibitor can effectively inhibit the formation of PCDD/Fs in flue gas, introducing sulphurous compounds into the waste mix directly led to a high SO₂ emission level in flue gas. The correlation between NO₂ and PCDD/Fs formation could be explained as NO_x specifically NO₂ act as an oxidizing agent and participates in chemical reactions with organic compounds including those containing chlorine. These reactions may have led to the formation of PCDD/Fs.

Table 3 provides data correlation between the reduction in PCDD/F emissions and the concentrations of other gases when using AC. In the primary chamber, where PCDD/F emissions were higher, the O₂ level was 8%. This suggests more incomplete combustion, resulting in high PCDD/Fs emissions. Conversely, in the secondary chamber, when the temperature increased from 900°C to 1150°C and O₂ levels increased to 10%, PCDD/F emissions reduced from 109.5 ng/Nm³ to 10.89 ng/Nm³, indicating complete combustion. Thus, a decrease in PCDD/F emissions correlates with an increase in O₂ levels and temperatures. High NO_x concentrations of 60 ng/Nm³ were detected within the primary chamber. However, with an increase in temperature, these concentrations notably decreased to 25 ng/Nm³ within the secondary chamber. Interestingly, a discernible correlation emerged between the reduction in NO_x concentrations and a concurrent decrease in PCDD/F emissions. This suggests a potential relationship between the levels of NO_x and the emission of PCDD/F. Initially, a concentration of 40 ng/Nm³ of SO₂ was observed within the primary chamber. This high concentration suggests the potential for SO₂ to inhibit the formation of PCDD/Fs. However, in the secondary chamber where PCDD/F emissions were lower, the SO₂ concentration also decreased to 15 ng/Nm³. Therefore, it appears that as PCDD/F emissions decrease, there is a corresponding decrease in SO₂ concentration, hinting at a complex relationship between these pollutants. Higher HCl concentrations in both chambers correlate with lower PCDD/F emissions. This suggests that increased HCl concentrations may contribute to the reduction of PCDD/F emissions, possibly by influencing the partitioning of PCDD/Fs between gas and particle phases. Therefore, an increase in PCDD/F emissions correlates with an increase in HCl concentration. These correlations suggest complex interactions between combustion conditions, pollutant concentrations, and PCDD/F formation pathways during the incineration process.

Table 3. Correlation between PCDD/Fs formation with other flue gases

| Gas Compositions | Temperature (°C) | O ₂ % | SO ₂ (ng/Nm ³) | NO _x (ng/Nm ³) | HCl (ng/Nm ³) | PCDD/F (ng/Nm ³) |
|-------------------|------------------|------------------|---------------------------------------|---------------------------------------|---------------------------|------------------------------|
| Primary Chamber | 900 | 8 | 40 | 60 | 25 | 109.5 |
| Secondary Chamber | 1150 | 10 | 15 | 25 | 45 | 10.89 |

3.5 Spent activated carbon desorption

Spent AC was collected from bag filters after the injections and analyzed for PCDD/Fs concentrations. The desorption of PCDD/Fs from spent AC was a crucial step in the treatment

process, aiming to recover the adsorbent's efficiency while mitigating environmental risks. During the treatment of flue gases from waste incineration, AC served as an effective adsorbent for capturing of PCDD/Fs due to its high surface area and porosity. However, during the study it was found that,

through de novo synthesis, the AC became saturated with adsorbed PCDD/Fs, necessitating desorption to regenerate its adsorption capacity. While ensuring the safe disposal of the spent AC without posing any environmental risks, cost-saving measures were identified by returning the spent AC back to the primary chamber of the incinerator where temperatures ranged between 850°C and 900°C. At these elevated temperatures, the PCDD/Fs desorb from the surface of the AC and revert to gaseous form, allowing them to be safely incinerated along with the waste material. During the study, it was found that the desorption of PCDD/Fs from spent AC was influenced by various factors, including temperature, residence time, and the chemical nature of the adsorbent and contaminants. Studies such as Wang et al. [21], Gerasimov et al. [32], Cerasa et al. [42], and Zhang et al. [43] have shown that desorption efficiency depends on the degree of chlorination of the PCDD/F congeners, with higher chlorinated congeners requiring higher temperatures for desorption due to their lower vapor pressures.

Furthermore, the desorption process was carefully controlled to prevent the release of PCDD/Fs back to gaseous phase into the environment. PCDD/Fs concentration was measured at 150 ng/Nm³ before desorption process indicating a significant presence of PCDD/Fs compounds in the spent AC. After undergoing desorption treatment, the concentration decreased substantially to 0.13 ng/Nm³, highlighting the effectiveness of the desorption process in removing PCDD/Fs from the spent AC. It's important to note that desorption is not only aimed at reducing the concentration of PCDD/Fs but also serves to regenerate the AC, allowing it to be reused for further adsorption cycles, in this case in the landfill site where AC and ash was disposed. While the potential effects of utilizing treated AC in a landfill setting were not scientifically assessed in the present study, landfills often produce unpleasant odors due to the decomposition of organic waste. According to Mallick et al. [44] treated AC can be used as a cover material or integrated into landfill liners to help mitigate odors by adsorbing volatile organic compounds (VOCs) and other malodorous substances. Landfills also pose a risk of contaminating groundwater with leachate containing harmful substances. Treated AC can be used in permeable reactive barriers (PRBs) or as part of groundwater treatment systems to adsorb contaminants and prevent their migration into groundwater resources [45]. Talebi et al. [46] added that landfills are significant sources of methane, a potent greenhouse gas. While treated AC may not directly capture methane, it can be utilized in methane oxidation systems or combined with other materials to enhance methane capture and oxidation efficiency, thereby reducing greenhouse gas emissions from landfills.

4. CONCLUSION

The objective of this study was to assess the effectiveness of AC injections in absorbing PCDD/Fs from the flue gas emissions in the MSWI. The study employed a comprehensive approach, combining experimental investigations with mathematical modeling to assess the impact of AC injection on PCDD/F emissions. Laboratory-scale experiments were conducted to evaluate the efficiency of AC in adsorbing PCDD/F congeners under controlled conditions, providing

fundamental data for understanding the mechanisms involved. Furthermore, rigorous sample preparation, calibration, and validation procedures were implemented to ensure the accuracy and reliability of results, particularly in environmental assessments and regulatory compliance. Based on the comprehensive analysis of the findings obtained from the current study on the mitigation of PCDD/F emissions from waste incineration processes through AC injections, several key conclusions can be drawn. Firstly, the current study demonstrated that AC injection technology effectively reduces PCDD/F emissions from waste incineration processes, achieving significant removal efficiencies of up to 85%. The technology offered a cost-effective and relatively straightforward approach to controlling PCDD/F emissions, making it promising for application in various waste incineration facilities. Secondly, the effectiveness of AC injections in reducing PCDD/F emissions was influenced by various factors, including the injection rates, chemical properties of PCDD/F congeners, temperature, residence time, and the presence of other flue gases. Higher injection rates and temperatures generally resulted in improved removal efficiencies, while certain congeners exhibited different responses to AC treatment. Thirdly, the study highlighted the complex interplay between PCDD/F congeners, waste composition, incineration conditions, and AC adsorption selectivity. PCDF congeners generally responded better to AC treatment compared to PCDD congeners, and highly chlorinated congeners exhibited lower removal rates compared to less chlorinated ones. Additionally, the desorption of PCDD/Fs from spent AC was a crucial step in the treatment process, aiming to regenerate the adsorbent's efficiency while mitigating environmental risks. The desorption process, influenced by factors such as temperature and residence time, effectively removes PCDD/Fs from spent AC, allowing for its safe disposal or potential reuse.

However, despite the promise shown by AC treatment in reducing PCDD/F emissions, further optimization and technological advancements such as the selective catalytic reduction (SCR) systems, entrained flow reactors, and the application of electron beam systems are necessary to meet stringent regulatory standards. Previous studies have identified inconsistencies in the impact of AC on the removal efficiency of PCDD/Fs, and the current study has validated those findings. To address these inconsistencies and improve the understanding of AC's impact on PCDD/Fs removal efficiency, future research could focus on standardizing experimental protocols, characterizing AC properties comprehensively, accounting for interfering compounds, employing robust sampling and analytical techniques, and conducting systematic comparative studies under controlled conditions. Additionally, advanced modeling approaches could be utilized to simulate and optimize the performance of AC-based air pollution control systems for PCDD/Fs mitigation. In conclusion, while AC injection technology shows promise for mitigating PCDD/F emissions from waste incineration processes, further optimization and complementary control measures may be necessary to meet stringent regulatory requirements. Future research should focus on optimizing AC injection parameters, understanding the behavior of PCDD/F congeners during AC treatment, and exploring the potential reuse of spent AC in landfill applications or groundwater treatment systems.

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