Agronomy Research **16**(S1), 1052–1061, 2018
https://doi.org/10.15159/AR.18.031

Stabilization of persistent organic pollutants (POP) in flue gases in a biological waste incinerating facility

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Abstract. The article focuses on a technology of flue gases stabilization in biological and hospital waste incinerating facility. Hazardous particles and compounds formed in flue gas during the process of thermal waste degradation need to be stabilized according to an enacted legislation. The aim of the research is to examine technological process of polychlorinated dibenzodioxins and polychlorinated dibenzofurans (PCDD/F) elimination in flue gas. PCDD/F is group of persistent organic pollutants which are resistant to environmental degradation and are highly toxic for the environment in very small amounts. Experiments were performed in semi operation hospital waste incinerating plant with heating power 1.5 MW and capacity 250 kg h^{-1} of incinerating material. It was observed that formation of PCDD/F exponentially grows with increasing concentration of chlorides in the flue gas, but formation varies based on type of congener. The highest concentration was measured for HxDF, 17,522 pg Nm^{-3} and HpDF, 16,334 pg Nm^{-3} at chloride concentration of 867 mg Nm^{-3}. However, concentration of PCDD congeners didn´t exceeded 4,000 pg Nm^{-3} for the same level of chlorides in flue gas. Two types of activated carbon Chezacarb and NORIT were tested for capability to stabilize PCDD/F in flue gas. Results show that refining effect for both sorbent are very similar. It was observed that 0.1 g of activated carbon should be applied per 1 Nm^3 of flue gas in regard to reduce 3–4 ng Nm^{-3} PCDD/F under required emission limit.

Key words: PCDD, PCDF, hospital waste, activated carbon.

INTRODUCTION

Elimination of hazardous particles and compounds in flue gas can be done by several methods such as a wet, semi-dry and dry process (Wey et al., 2001). Dry refining technology is based on a sorbent injection that is in a form of finely crushed dry powder. The agent is sprayed directly into the flue gas flow (Allen et al., 2001) or mixed with waste going into the reactor (Cunill et al., 1991). Neutral compounds are formed after reaction with injected sorbent and captured on solid particles separator subsequently. There is a vast range of commercially produced sorbents that can be used for stabilization of different particles with different efficiency. Sodium and calcium based sorbents can effectively eliminate sulphur dioxide and hydrogen chloride (Jirsa & Malaťák, 2017), but for activated carbon based sorbent seems to be effective agent for persistent organic pollutants stabilization (Gunes et al., 2015).

Hydrogen chloride and small amount of chlorinated radicals can form during incineration of material that contains chemically bonded chlorine (Phan et al., 2014). The combustion runs in oxidation atmosphere in presence of catalytically active particles of ash such as Cu^{2+}. It is known as a Deacon´s process (Hisham & Benson, 1995). Under such condition, hydrogen chloride can transform to chlorine that can reacts with organic radicals and persistent Polychlorinated dibenzodioxins (PCDD) and Polychlorinated dibenzofurans (PCDF) can form (Phan et al., 2014).

PCDD/F occurs while incineration of substances that contain chlorine (Zhou et al., 2015). It is highly toxic for the environment in very small amounts. Production of PCDD/F in flue gases grows with increasing amount of hydrogen chloride and other chlorine element in flue gas (Van Caneghem et al., 2012). Its formation occurs at temperature range from 250 to 600 °C (Stanmore, 2004). Emission limits for PCDD/F is 0.1 ng I-TEF m^{-3} at standard conditions.

This paper follows up on research of the dry sorption refining technology in small biological and medical waste incinerating plant with heating power under 5 MW (Jirsa & Malaťák, 2017). The aim of this study is to determine a formation of PCDD/F in flue gas during medical waste incineration in relation with concentration of chlorides. Further task of the study is to examine the effect of calcium hydroxide on PCDD/F elimination. Above all, it is determination of sorption efficiency of activated carbon-based sorbents Chezacarb and NORIT to reduce PCDD/F concentration in flue gas.

MATERIALS AND METHODS

Experimental unit and technology

All experimental tests were conducted in semi-operation conditions of incinerating plant for hospital waste degradation in Strakonice, Czech Republic. It is a small-scale facility with output of approximately 1,400–1,500 t processed waste annually. The facility consists of a pyrolysis furnace Hoval – Schiestl GG 24, thermo-reactor TR 24 and a steam boiler THD IV 0580 with heating power 1,508 kW.

The incinerating plant operates discontinuously, waste is dosed into the furnace every 15 minutes with load of 60–65 kg. The input material is a classified as a biological and hospital waste. It is described with following codes according to waste catalogue: 180102, 180103, 180104, 200301, 150202, 150110 (European Waste Catalogue). The elemental characteristic of processed waste is in Table 1.

Table 1. Hospital waste characteristics

	Share of component, %	H_2O, %	Ash, %	Cl, %	F, %	S, %	Heating value, MJ kg^{-1}
Textile	29	6.6	14.2	13.2	1.8	9.5	15.1
Paper	24	5.8	5	0.5	-	4.2	15.4
Plastic	23	0.1	0.5	18	3	6.7	24.8
Biological component	14	32.1	18.6	5.3	2.1	12	11.4
Solvent, oil	10	0.6	0.8	16.1	1.3	21.3	39.6
Weighted average		**7.9**	**8.1**	**10.4**	**1.6**	**9.1**	**19.3**

Temperature in the furnace chamber is set at range 900–950 °C. Two natural gas burners automatically control temperature at required level. Pyrolysis gas forms by waste carbonization in the furnace chamber that is subsequently mixed with oxygen and burned in thermo-reactor at the temperature 1,100–1,150 °C. Formed flue gas flows through a boiler where the temperature is reduced to 200–250 °C. Reduced temperature of flue gas allows to inject a sorbent into the system. Hence the device for sorbent addition is placed after the boiler. Flue gas followingly passes through 8 m long contactor-pipe labyrinth which enhance amount of collisions between sorbent and flue gas particles. Next, flue gas temperature is reduced to 130–135 °C in heat exchanger after which second 17 m long contactor-pipe labyrinth is placed. All solid particles involving hazardous compounds and sorbents injected into the system are captured on a tied filter subsequently. The technology diagram of the incinerating facility is given on the Fig. 1.

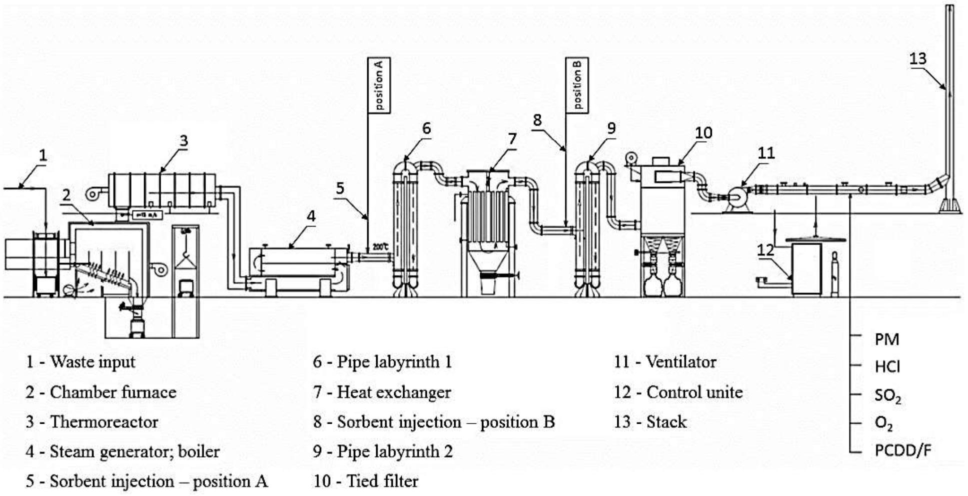

Figure 1. Facility technology diagram (Jirsa & Malaťák, 2017).

Measuring of emissions in flue gas was taken according to appropriate legislative (Act No. 201/2012 Coll.). The measuring point is placed at the horizontal flue pipe with diameter 355 mm after a ventilator according to ISO 9096 and EN 13284-1.

Within the experimental testing following properties and concentrations are determined according to international technical standards. It is specifically monitored: concentration of particulate matter (PM), hydrogen chloride (HCl), oxygen (O_2) and polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) according to following technical standards:

- Gravimetric determination of the mass concentration of particulate matter (ISO 9096 EN 13284-1);
- Determining flow velocity and volume flow (ISO 10780);
- Determination of moisture in the gas pipeline (EN 14790);
- Paramagnetic determination of oxygen (EN 14789);
- Photometric determination of the mass concentrations in inorganic chloride compounds (EN 1911);
- Determination of the organic persistent substances mass, calculated from the measured values (PCDD/PCDF) (EN 1948, 1-4).

Methodology of PCDD/F determination according to EN 1948, 1-4

Equipment was rinsed with acetone and toluene before the samples were taken. PUF and glass filter were extracted with toluene and dried. PUF and glass filter was marked with sampling standards for yield determination and blinded test was done then.

PCDD/F samples were determined in the laboratory with addition of C^{13} internal standards (extraction and spraying standards) and determined on the GC-MS device. The yield of sampling standards was determined based on extraction standards. The results are given as an International toxicity equivalence factor I-TEF, see the Table 2.

Table 2. Toxic equivalency factor (Schecter et al., 2006)

Congener	I-TEF value	Congener	I-TEF value
Dibenzo-p-dioxins (PCDDs)		**Dibenzofurans (PCDFs)**	
2,3,7,8-TCDD	1	2,3,7,8-TCDF	0.1
1,2,3,7,8-PeCDD	0.5	1,2,3,7,8-PeCDF	0.05
		2,3,4,7,8-PeCDF	0.5
1,2,3,4,7,8-HxCDD	0.1	1,2,3,4,7,8-HxCDF	0.1
1,2,3,6,7,8-HxCDD	0.1	1,2,3,6,7,8-HxCDF	0.1
1,2,3,7,8,9-HxCDD	0.1	1,2,3,7,8,9-HxCDF	0.1
		2,3,4,6,7,8-HxCDF	0.1
1,2,3,4,6,7,8-HpCDD	0.01	1,2,3,4,6,7,8-HpCDF	0.01
		1,2,3,4,7,8,9-HpCDF	0.01
OCDD	0.001	OCDF	0.001

All volumes, weights of combustion air and emission concentration are given for normal conditions, i.e., at $t = 0$ °C, $p = 101.325$ kPa and the reference oxygen content in the flue gases $O_r = 11\%$.

Used carbon based sorbents

Chezacarb (CHZC) is commercial name for carbon based sorbent produced by Unipetrol a.s. It is produced during hydrogen production by partial oxidation of petroleum and petrochemical raw materials. Chezacarb is characterized by high level of specific surface area, above 1,000 $m^2\ g^{-1}$ with high capacity and high porosity, more than 3.5 $cm^3\ g^{-1}$. It is effective in PCDD/F and PCB elimination. Small particles around 20 mm are able to cluster around pollutants and binding them irreversibly (Unipetrol, n.d.).

NORIT GL 50 is commercial name for carbon based sorbent produced by Cabot Corporation. It is a powdered steam activated carbon with an extra fine particle size that is suitable for the removal of dioxins, mercury and other contaminant traces from flue gases (Cabot Corporation, n.d.).

RESULTS AND DISCUSSION

Effect of chlorine in flue gas on PCDD/F and distribution of congeners

The dependence of PCDD/F formation on hydrogen chloride concentration in flue gas was observed. The characteristics of the biological waste meet the fuel quality requirements. An average calorific value corresponds to wood (Malaťák & Kučera, 2013) or vegetable matter (Malaťák & Bradna, 2014). The concentration of hydrogen chloride was changed by mixing 100 kg of crushed PET bottles and 3 litres of 4.6% solution NaClO with incinerated waste before dosage to the burning chamber.

The measuring point was placed in front of the fabric filter. Flue gas temperature was 125–130 °C and volume of oxygen was 8.09–8.83%. The dependence of PCDD/F formation based on hydrogen chloride concentration in flue gas is shown in Figs 2, 3.

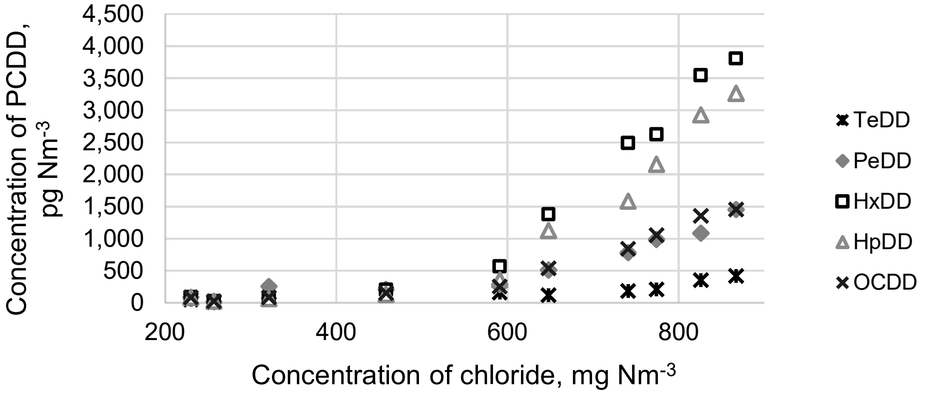

Figure 2. Dependence of PCDD individual congeneres concentration on chlorides concentration in flue gas.

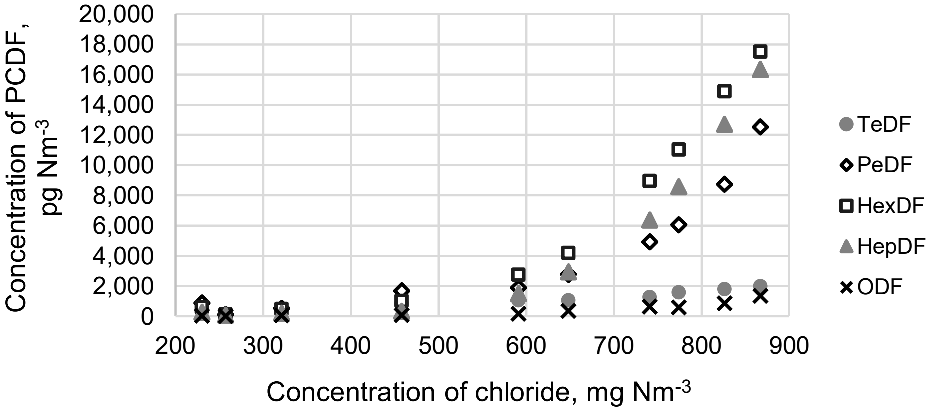

Figure 3. Dependence of PCDF individual congeneres concentration on chlorides concentration in flue gas.

The results show that the formation of both PCDD/F exponentially grows with the concentration of chlorides in the flue gas. However, the formation can vary based on type of congener. The distribution of concentration for different PCDD/F congeners are shown in Figs 4, 5.

It is obvious that the concentration of polychlorinated dibenzofurans is almost one order of magnitude higher that the concentration of polychlorinated dibenzodioxins. At chloride concentration of 458 mg Nm^{-3} formation of TeDD, PeDD, HxDD, HpDD and OCDD do not vary significantly. All the congeners formed at similar level between 150–200 pg Nm^{-3}. However, at chloride concentration of 867 mg Nm^{-3} formation of TeDD, PeDD, HxDD, HpDD and OCDD difers noticeably. Concentration of congener with the lowest number of chlorine atoms, TeDD does not grow with increasing concentration of chlorides in flue gas. Concentration of PeDD and OCDD slightly increases with chlorides in flue gas. Concentration of HxDD and HpDD grows the most significantly with increasing concentration of chlorides in flue gas. The maximum concentration of is 3,811 pg Nm^{-3} for HxDD and 3,266 pg Nm^{-3} for HpDD at chloride concentration 867 mg Nm^{-3}.

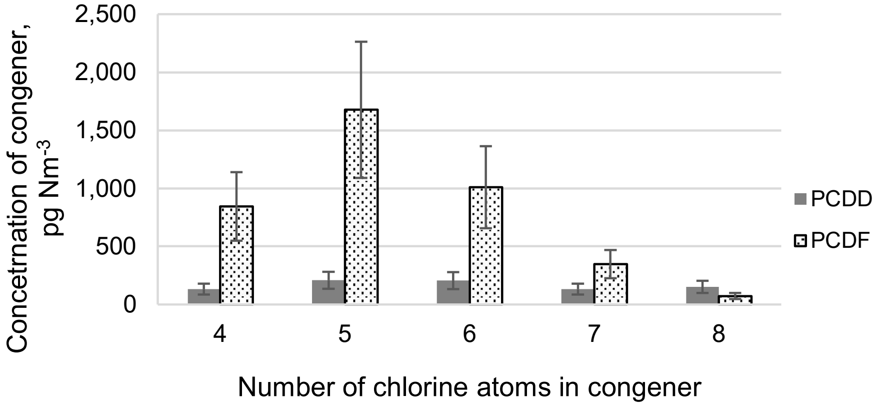

Figure 4. Concentration of persistent compounds in flue gases with content of chlorine equal to 458 mg Nm^{-3}.

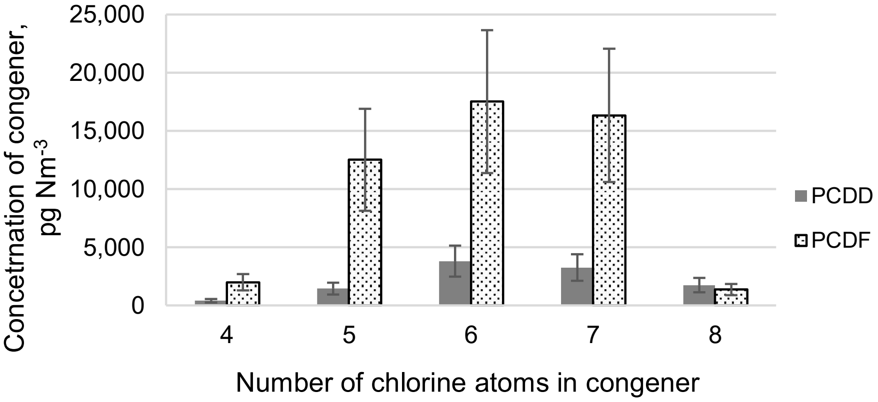

Figure 5. Concentration of persistent compounds in flue gases with content of chlorine equal to 867 mg Nm^{-3}.

Compared to that formation of polychlorinated dibenzofurans conducts differently. At chloride concentration of 458 mg Nm^{-3} TeDF, PeDF, HxDF form noticably more than HpDF and OCDF. The highest concentration 1,677 pg Nm^{-3} is masured for PeDF. However, concentration of OCDF is only 77 pg Nm^{-3}. With increasing concentration of chloride in flue gas congeners with 5, 6 and 7 chlorine atoms form the mostly. Maximum concentration of HxDF is 17,522 pg Nm^{-3} at chloride concentration of 867 mg Nm^{-3}. Similarly, concentration of the second most present congener, HpDF is 16,334 pg Nm^{-3} at the same level of chlorides in flue gas. On th other hand, concentration of TeDF and OCDF does not grow as signiciantly as concentration of PeDF, HxDF and HpDF with increasing concentration of chlorides in flue gas, but still quite significanly compare to PCDD. The maximum concentration is 1,966 pg Nm^{-3} for TeDF and 1,361 pg Nm^{-3} for OCDF at chloride concentration of 867 mg Nm^{-3}. Very similar results were published for industrial incinerators (Chi et al., 2006) and for municipal waste incinerators (Kim et al. 2007).

Noticeable slope change of concentration fuction for individual congeners is around 600 mg Nm^{-3} of chlorides in flue gas when concentration of HxDD, HpDD and PeDF, HxDF, HpDF start growing dramatically compared to other PCDD and PCDF congeners. HxDF, HpDF have the major presence in flue gas.

Effect of $Ca(OH)_2$ on PCDD/F concentration reduction in flue gas

The influence of calcium hydroxide on concentration of PCDD/F in flue gas was measured. Calcium hydroxide was injected into the flue gas flow at rate of 5 kg per hour. The sorbent was dosed after a cooler where the temperature of flue gas was 225 ± 3 °C. The mean volume of flue gas was 3,296 $Nm^3 h^{-1}$ and mean volume of O_2 was 10.3 ± 0.95%. The concentration of hydrogen chloride decreased from original concentration of 458 mg Nm^{-3} to 230 mg Nm^{-3}. The reduction of individual congeners PCDD/F are shown in Table 3, 4.

Table 3. Effect of $Ca(OH)_2$ on PCDD concentration

PCDD	HCl	TeDD	PeDD	HxDD	HpDD	OCDD	Σ PCDD
I-TEF	mg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}
Initial concentration	458	133	208	206	133	152	832
Concentration when 5 kg $Ca(OH)_2$ dosed	230	51	86	94	85	87	403
Reduction of congener concentration		61.7%	58.7%	54.4%	36.1%	42.8%	51.6%

Individual congener reacts on calcium hydroxide addition to the system differently. The major reduction 61.7% was observed regarding TeDD. With growing order of chlorine atoms, the reduction slowly decreases. The mean decrease of all PCDD congeners is 51.6%.

Table 4. Effect of $Ca(OH)_2$ on PCDF concentration

PCDF	HCl	TeDF	PeDF	HexDF	HepDF	OCDF	Σ PCDF
I-TEF	mg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}	pg Nm^{-3}
Initial concentration	458	844	1,677	1,010	348	73	3,952
Concentration when 5 kg $Ca(OH)_2$ dosed	230	406	883	612	263	33	2,197
Reduction of congener concentration		51.9%	47.3%	39.4%	24.4%	54.8%	44.4%

Similar effect was observed in regards with individual PCDF congeners as well. Maximum reduction 51.9% was seen with decreased concentration of TeDF. With increasing number of chlorine atoms in individual congener effect of $Ca(OH)_2$ on subsequent PCDF reduction descened. The mean decrease of all PCDF congeners is 44.4% and 45.7% for both PCDD and PCDF together. The result shows that calcium hydroxide reduces the concentration of hydrogen chloride flue gas and subsequently reduces concentration of PCDD/F in flue gas.

Gunes similarly published that the lowest removal efficiency for particle phase was found to be 80% for TeDF while the highest removal efficiency found to be 100% for TeDD, PeDD and PeDF. Particle phase removal efficiencies for the rest of congeners were observed to range from 97–99% (Gunes et al., 2015). Lower chlorinated congeners have higher vapor pressures. So, they tend to go into gas phase and hence, have higher possibility of adsorption on activated carbon (Chi et al., 2006). On the other hand, higher chlorinated congeners tend to be present in particle phase due to their lower vapor pressures. These congeners are separated from gas stream after their adsorption onto activated carbon particles or their association with lime particles, consequently, filtration of the particles through bag house filter (Gunes et al., 2015).

Effect of activated carbon on PCDD/F concentration reduction in flue gas

Concentration of PCDD/F in flue gas can be decreased by calcium hydroxide that is usually used for reduction of acidic compounds in flue gas. However, the effect is not sufficient according to appropriate emission limits. Activated carbon is usually used for elimination of persistent organic pollutants in small incinerating facilities.

First measurement of PCCD/F concentration in flue gases was taken when neither sorbents nor active carbon was added into a system. It represents the reference point for evaluation. The volume of glue gas flow was 3,071 Nm^3 hr^{-1}.

Next 3.5 kg of calcium hydroxide per hour was injected into a flue gas stream after the heat exchanger (position A). The temperature of gases was 230 °C at dosing point. Calcium hydroxide can partially reduce concentration of PCDD/F by stabilizing chlorides in flue gas as it is shown in Table 4, 5. However, it cannot reduce PCDD/F at required level given by emission limits (Act No. 201/2012 Coll.).

Hence, activated carbon was examined on efficiency to diminish PCDD/F concentration to the acceptable level. There were two carbon based sorbents selected Chezacarb and NORIT, both crashed in high speed mill crusher to fine particles with size smaller than 5 µm. The dosing place had to be selected with regard to a flue gas temperature because there was a considerable content of oxygen. Dosing point was placed after a cooler (position B) where the temperature was 120 °C. Both sorbents Chezacarb and NORIT were dosed into the flue gas flow at rate of 300 g hr^{-1} and 400 g hr^{-1}. The results are shown in Table 5.

Table 5. Stabilization effect of activated carbon on PCDD/F concetration

	Reference emissions No sorbents added	$Ca(OH)_2$ 3.5 kg hr^{-1}	Chezacarb 300 g hr^{-1}	Chezacarb 400 g hr^{-1}	NORIT 300 g hr^{-1}	NORIT 400 g hr^{-1}
O_2	% vol.					
	8.58± 1.24	8.76± 1.28	8.45± 1.26	8.94± 1.48	8.65± 1.28	8.67 ± 1.30
Flue gas flow	Nm^3 hr^{-1}					
	3,071	3,208	3,710	4,100	3,890	4,020
Concentration of PCCD/DF	ng I-TEF Nm^{-3}					
	4.852	3.187	0.487	0.079	0.522	0.081

Result shows that both Chezacarb and NORIT are efficient to reduce concentration of persistent compounds in flue gases. Experiments were executed under the same conditions. All instruments and methodology were identical. Results are very similar, it is not possible to prefer one or the other sorbent based on sorption effectiveness. The results show that in order to satisfy emission limits for persistent compound approximately 0.1 g of activated carbon should be applied per 1 Nm^3 of flue gases with concentration 3–4 ng Nm^{-3} PCDD/F. Other researches published similar results. Abad stated that 92–96% removal efficiency can be achieved when 100 g Nm^{-3} of activated carbon is applied (Abad et al., 2003). Kim published that removal efficiency over 98% can be reached when 50 mg Nm^{-3} of activated carbon is injected into a system with dual bag filter, but 200 mg Nm^{-3} of activated carbon is necessary with a single filter applied (Kim et al., 2007).

CONCLUSIONS

The paper deals with formation and stabilization of PCDD/F during a process of biological and hospital waste thermal degradation. The experiments were conducted in a semi operation hospital waste incinerating plant with heating power 1.5 MW. Formation of PCDD/F exponentially grows with increasing concentration of chlorides in the flue gas, but formation varies based on type of congener. Concentration of PCDF is almost one order of magnitude higher that the concentration of PCDD. HxDF and HpDF reach the highest concentration 17,522 pg Nm^{-3} and 16,334 pg Nm^{-3} at chloride concentration 867 mg Nm^{-3}. However, concentration PCDD congeners does not exceed 4,000 pg Nm^{-3} at the same level of chlorides in flue gas. Calcium hydroxide dosed into the flue gas flow can decrease level of hydrogen chlorine and subsequently eliminate concentration of PCDD/F with the mean reduction of 45.7% at rate 5 kg h^{-1} of $Ca(OH)_2$ dosed into the flue gas flow. However, the effect of PCDD/F elimination by $Ca(OH)_2$ is not sufficient per emission limits. Activated carbon based sorbent is suitable to do so. Both examined carbon based sorbents commercially known as Chezacarb and NORIT achieve the same efficiency of PCDD/F stabilization in flue gas. It is not possible to prefer one or the other sorbent based only on sorption properties. In regards not to exceed emission limits of PCDD/F exposure, approximately 0.1 g of activated carbon must be applied per 1 Nm^3 of flue gases with total PCDD/F concentration of 3–4 ng Nm^{-3}. Carbon based sorbent is a vital supplement for PCDD/F elimination in dry sorption based refining technology in small scale waste incinerators.

ACKNOWLEDGEMENTS. The article was financially supported by the Internal Grant Agency of the Czech University of Life Sciences Prague, Czech Republic, Grant No. 20173002.

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