

## DIOXIN EMISSIONS FROM COAL COMBUSTION IN DOMESTIC STOVE: FORMATION IN THE CHIMNEY AND COAL CHLORINE CONTENT INFLUENCE

by

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*Combustion experiments conducted in domestic stove burning hard coal demonstrated a predominant influence of the coal chlorine content on the PCDD/F emissions, together with a pronounced effect of the flue gas temperature. PCDD/F concentrations of over 100 ng TEQ/m<sup>3</sup>, three orders of magnitude higher than in a modern waste incinerator, were measured in the flue gases of a domestic stove when combusting high chlorine coal (0.31%). The PCDD/F concentrations in the flue gases dropped below 0,5 ng TEQ/m<sup>3</sup>, when low chlorine coal (0.07%) was used. When low chlorine coal was impregnated with NaCl to obtain 0.38% chlorine content, the emission of the PCDD/F increased by two orders of magnitude. Pronounced non-linearity of the PCDD/F concentrations related to chlorine content in the coal was observed. The combustion of the high chlorine coal yielded PCDD/F concentrations in flue gases one order of magnitude lower in a fan cooled chimney when compared to an insulated one, thus indicating formation in the chimney. The influence of flue gas temperature on the PCDD/F emissions was less pronounced when burning low chlorine coal. The predominant pathway of the PCDD/F emissions is via flue gases, 99% of the TEQ in the case of the high chlorine coal for insulated chimney.*

Key words: *dioxins, furans, PCDD/F, stove, coal, combustion, chlorine, formation*

### Introduction

Polychlorinated dibenzo p-dioxins and furans (PCDD/F), often referred to as dioxins, are a group of structurally similar halogenated aromatic compounds. They are not produced intentionally, but they are formed in trace levels at elevated temperatures where carbon, hydrogen and chlorine containing compounds are present. The major sources of the PCDD/F are combustion and some specific chemical processes. There are 210 different PCDD/F compounds named congeners, differentiating on the level of chlorination and position of the chlorine atoms in the

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molecule. Among those, 17 congeners were attributed toxic properties. The summary toxicity of these PCDD/F is usually expressed as toxic equivalent (TEQ), where different PCDD/F congeners content is summarized, weighted by their relative toxicity compared to the most toxic 2, 3, 7, 8-tetrachlorodibenzodioxin. PCDD/F are among the most toxic compounds known. They are persistent in the environment, bio accumulate and bio magnify in the food chain. PCDD/Fs and their impact on human health are of great concern for scientists, policy makers and general public [1].

Small combustion installations significantly influence air quality in Europe. In urban areas combustion of wood and coal for domestic heating represents one of the mayor sources of particulate matter [2]. Solid fuel combustion in household appliances is also estimated to be the dominant source of the PCDD/F emissions into the air in the European Union [3]. Field studies of the spatial and temporal variations of the PCDD/F concentrations in the atmosphere [4, 5] in combination with typical congener profiles [6] suggest significant contribution of small solid fuel fired domestic combustion appliances to the elevated concentrations of the PCDD/F in atmosphere.

Combustion of virgin wood in small combustion installations typically leads to the PCDD/F concentrations in the flue gases below 1 ng TEQ/m<sup>3</sup>. Elevated concentrations of PCDD/F above 1 ng/TEQ, *i. e.* ten times higher than in modern waste incineration plants, were measured only when chemical contaminated wood was combusted [7]. Most experiments with coal combustion in domestic stoves did not show elevated concentrations PCDD/F in flue gases [8] either. However some authors measured extreme concentrations of PCDD/F, up to more than two orders of magnitude higher than those in modern waste incinerators [9, 10]. Extreme PCDD/F concentrations are deemed to be linked with high chlorine content of the coal, although the studies [9, 10] did not include data on the chlorine content of the coal. In a previous study [11] formation of the PCDD/F in the chimney was also identified, which further increases the complexity of the interpretation of the existing literature findings with regard to the influence of the chlorine content of coal.

In this paper the results from a series of controlled laboratory experiments are presented. The effect of different chlorine content of coal as well as the influence of different chimney configurations on PCDD/F in flue gases, bottom ash and chimney soot is shown.

## Experimental

### *Combustion experiments*

A commercial stove with nominal thermal capacity of 4 kW was used for the combustion experiments. The stove was of advanced construction with S-draught combustion [12] and intended for hard and brown coal of grain size between 10 and 50 mm. The combustion was controlled by regulation of the primary and secondary combustion air as well as by regulation of the flap at the top of the chimney thus influencing the natural draught.

A stainless steel duct of 136 mm in diameter and a height of 13 m served as chimney. Four chimney configurations were used in order to obtain different temperatures of the flue gases in the chimney:

- the chimney duct was insulated in the whole length with the mineral wool with 25 mm thickness,
- the chimney duct was left non-insulated,
- the heat exchanger was inserted in-between the stove outlet and the non-insulated chimney,
- the heat exchanger was forcedly cooled by a frequency controlled fan to decrease the temperature of the flue gases to 200 °C at the heat exchanger outlet.

The heat exchanger, typically used in households to increase overall efficiency of stove by lowering the temperature of the flue gases, consisted of seven in parallel positioned steel tubes with diameter of 50 mm. The total length of the heat exchanger was 0.6 m.

Each combustion cycle consisted of two phases. During the initial phase, which lasted one hour, basic firebed was prepared. It served for the ignition of the main batch of 5 kg coal added at the beginning of the operational phase. During the operational phase the complete main batch of the coal was combusted – its duration was determined by obtaining the same amount of the basic firebed on the stove grating as there was at the beginning. The operational phase lasted for 3 h 45 min. The emission measurements started when the main batch of coal was added and were continued during the whole operational phase. In this way we measured also the emissions during the ignition of the main batch of coal.

After the experiments applying a specific chimney/coal configuration, the chimney was thoroughly swept. The chimney soot was collected, weighted and stored. Bottom ash was collected after each combustion test.

Two types of the hard coal were selected for the experiments. Both were of Polish origin and available on the market for the household use. They were selected for their different chlorine content. One has the chlorine content of 0.31% and is referred as the high chlorine coal in this article, while the other having 0.074% of the chlorine is referred as the low chlorine coal (tab. 1).

**Table 1. Properties of the high and low chlorine coals**

Coal type	Higher heating value	Lower heating value	Moisture	Ash	Volatile organic compounds	Carbon	Hydrogen	Sulphur	Nitrogen	Chlorine
	H <sub>s</sub>	H <sub>i</sub>								
	[MJ/kg]		[%]							
High Cl	31.2	30.0	3.8	4.3	32.45	79.8	4.65	0.32	1.26	0.313
Low Cl	26.42	25.3	4.5	15.7	31.7	64.9	4.27	0.46	1.08	0.074

In order to vary the chlorine content of the fuel high and low chlorine coal were mixed. In addition a batch of low chlorine coal was immersed in a saturated NaCl solution and subsequently dried. The chlorine content of the NaCl impregnated coal was 0.378%.

#### *Emission measurements*

The PCDD/F sampling was performed according to the standard EN 1948 – using a cooled probe [13, 14]. Due to the low velocity of the flue gases in the chimney its direct measurement by the Prandtl-Pitot tube was not accurate. The sampling was therefore performed at a constant speed 1.2 m/s at the nozzle. This was the average velocity of the flue gases in the chimney and was determined from the calculated flue gas volume based on the elemental composition of the coal and its consumption as well as the composition of the flue gases. Because of the high particulate matter (PM) content in the flue gases and consequent possibility of blocking the filter the sampling volume was reduced to approximately 1 m<sup>3</sup> per combustion test. Extraction, clean up and analysis of PCDD/F was performed in line with the EN 1948 standard [13, 14]. Identification and quantification of the PCDD/F were done via isotope dilution using a high resolution gas chromatography/high resolution mass spectrometer (HRGC/HRMS) at a resolution of 10000. The TEQ PCDD/F levels in this article are presented in International Toxic Equivalent metrics [13, 14].

The PM was measured gravimetrically. Like for PCDD/F, the sampling of the PM was done at the constant speed. In this was we ensured that the sampling was on the average performed isokinetically. The H<sub>2</sub>O, CO, CO<sub>2</sub>, NO, SO<sub>2</sub>, and HCl concentrations in the flue gases were measured by extractive Fourier transformation infrared spectrometry. Volatile organic compounds (VOC) in the flue gases were measured with a flame ionisation detector and the oxygen content via paramagnetic alternating pressure method.

The upper sampling point for the PCDD/F was positioned at 6.3 m from the stove outlet to the chimney. The lower sampling point for the PCDD/F, which was used for the purpose of indication of the possible PCDD/F formation in the chimney, was 0.6 m from the stove outlet. The temperature of the flue gases in the chimney was measured at six points. The distances of the temperature measuring points from the stove outlet are given in tab. 2.

**Table 2. Average temperatures of the flue gases and emission factors for different chimney configurations**

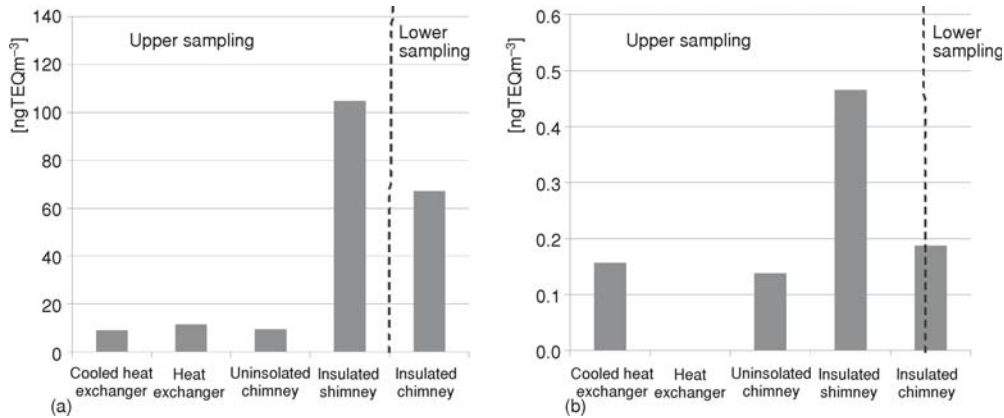
Coal type	Chimney configuration	T1	T2	T3	T4	T5	T6	CO	SO <sub>2</sub>	NO <sub>x</sub>	VOC	HCl	PM	PCDD/F
		[°C]						[kgf <sup>-1</sup> ]						µgTEQ/t
High Cl	Insulated	419	385	353	317	288	264	15.2	2.8	3.5	8.1	2.6	3.9	1326
	Non-insulated	332	261	228	193	166	146	14.9	3.2	3.5	5.9	2.7	2.8	126
	Heat exchanger	230	202	172	154	136	121	17.0	2.7	3.1	6.6	2.2	3.8	131
	Cooled heat exchanger	190	162	143	124	110	99	21.9	2.7	3.3	8.8	2.3	4.4	112
Low Cl	Insulated	361	297	277	262	240	216	14.1	4.5	2.7	5.5	0.4	2.0	3.77
	Non-insulated	274	237	215	189	159	151	10.1	4.1	2.3	4.2	0.3	1.8	1.06
	Heat exchanger	235	200	180	155	135	122	11.3	4.6	2.8	4.0	0.4	1.7	–
	Cooled heat exchanger	178	157	143	126	105	103	13.6	4.4	2.8	5.6	0.3	1.8	1.35
Distance of measuring/sampling point from stove outlet [m]		0.7	1.8	3.1	4.2	5.3	6.4	4.7	4.7	4.7	4.7	4.7	6.9	6.4

The emission factors, *i. e.* emissions per mass of the coal were determined using the measured concentrations and the theoretical volume of the flue gases based on the coal analysis.

## Results and discussion

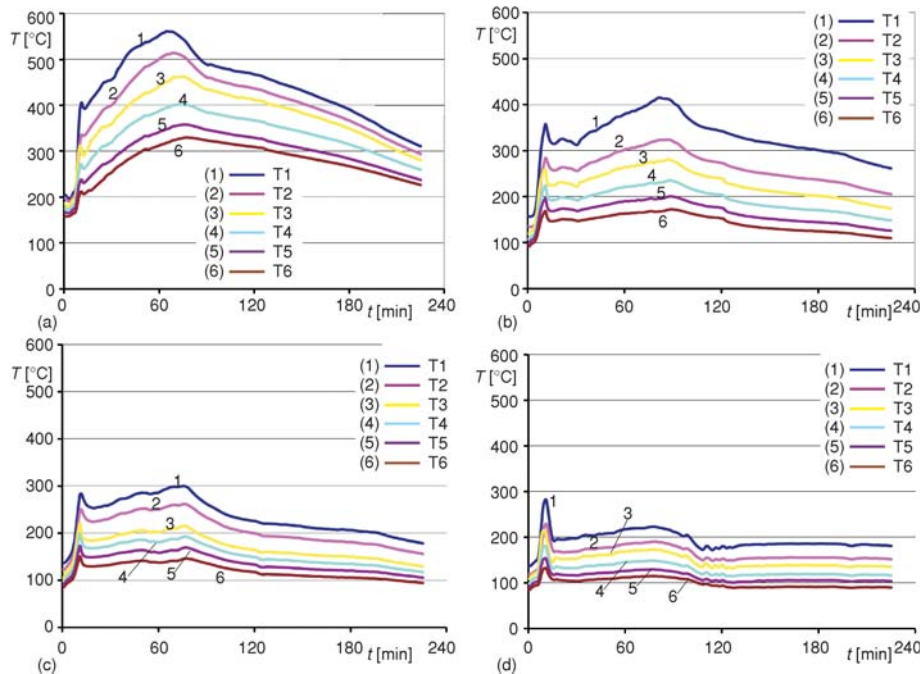
### *PCDD/F formation and chimney configuration*

The emissions of SO<sub>2</sub>, NO<sub>x</sub>, CO, VOC, and PM (tab. 2) were not dependent on the chimney configuration. This is an indication, that the combustion process, as expected, was not influenced by different chimney configurations. On the contrary, the PCDD/F concentrations measured in the flue gases reflect significant influence of the chimney configuration. The most pronounced influence was observed with high chlorine coal fig. 1(a), where there was a difference of one order of magnitude between concentrations of the PCDD/F measured in the insulated chimney and in other chimney configurations. In the case of low chlorine coal the difference in the PCDD/F concentrations measured with different chimney configurations was also observed, but was smaller than when high chlorine coal was used fig. 1(b). The PCDD/F levels for experiments with low chlorine coal and heat exchanger are not presented, because the sample was lost.



**Figure 1. PCDD/F concentrations in flue gases for different chimney configurations and sampling points with low chlorine (a) and high chlorine coal (b) recalculated to 11% oxygen concentration**

Different chimney configurations resulted in different temperature profiles of the flue gases in the chimney, however the time dependence was similar (fig. 2). A local temperature maximum was achieved ten minutes after the start of the beginning of the measuring cycle due to intensive devolatilisation of the coal. The absolute maximum of the temperature was reached after one hour, except when heat exchanger was cooled.



**Figure 2. Typical temperature profile of the flue gases in the chimney for various chimney configurations: (a) insulated chimney, (b) non-insulated chimney, (c) non-insulated chimney and heat exchanger, (d) non-insulated chimney and cooled heat exchanger. Positions of the temperature measuring points are given in tab. 1**

In a non-insulated chimney the differences in the PCDD/F levels at the upper sampling point were small for both types of coals, independently from whether a heat exchanger was in place. Also the cooling of the heat exchanger caused no remarkable variations. In contrast, the insulation of the chimney resulted in a pronounced rise in PCDD/F concentrations in the flue gases, in particular for the high-chlorine coal.

This leads as to the conclusion, that PCDD/F formation in the flue gases in the chimney is not pronounced in all chimney configurations without insulation. However in the case of high chlorine coal and the insulated chimney we estimate that approximately 90% of the PCDD/F were formed in the chimney.

Measurements of the PCDD/F concentrations at the lower sampling point do not include the PCDD/F emissions that are possibly formed in the chimney at the distances greater than 0.6 m from the stove outlet. Measurements of the PCDD/F concentrations at the lower sampling point were performed only with the insulated chimney. In the case of the high chlorine coal PCDD/F concentrations in the flue gases at the lower sampling point were 30% smaller than PCDD/F concentrations at the upper sampling point. This indicates, that when high chlorine coal is combusted and insulated chimney used PCDD/F mostly form in the part of the chimney near to the stove outlet. In the case of low chlorine coal the difference in PCDD/F concentrations between upper and lower sampling point was in the range of factor 2.

PCDD/F can be formed in catalysed reaction on the surface of the particles (heterogeneous reaction) or in the gas phase (homogenous reaction). The temperature window for the heterogeneous reaction is below 400 °C, while homogenous reaction starts at approximately 500 °C [15]. Based on our observation, that the PCDD/F were formed predominately in the insulated chimney and in the lower part of the chimney, it could be concluded, that the PCDD/F formation follows homogenous pathway. It should be noted herein, that only in the lower section of the insulated chimney temperatures above 500 °C, promoting a homogeneous reaction, were observed (fig. 2a).

#### *PCDD/F formation and chlorine content in coal*

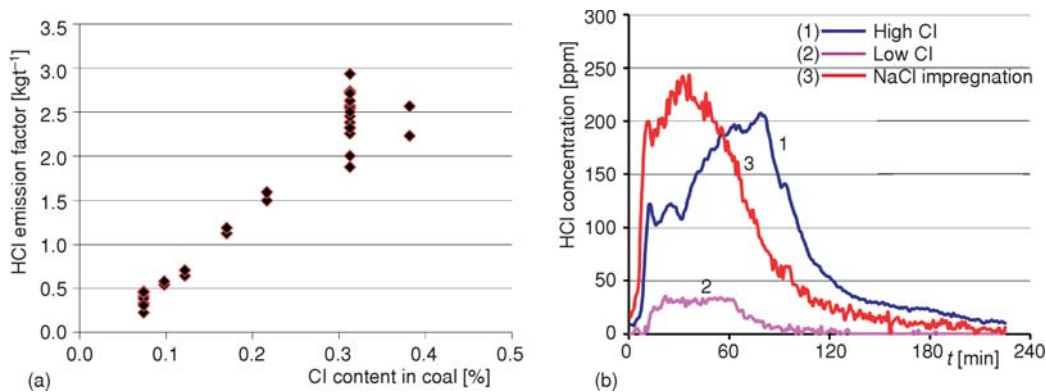
The results presented in the previous section show the significant difference in PCDD/F emissions when using high and low chlorine coal for all chimney configurations. In order to gain additional information on the coal chlorine content influence the experiments with coal mixtures and NaCl impregnated coal were performed. Those experiments were performed with insulated chimney configuration, because we consider, that this chimney configuration is the closest to the typical situation in households in respect of thermal properties.

Results showed pronounced and non-linear dependence of the PCDD/F emissions on the coal chlorine content. PCDD/F emissions up to 0.2% of chlorine in the coal exhibit small differences, but sharply increase at the higher concentrations. The lowest PCDD/F emissions were measured with low Cl coal. The highest PCDD/F emissions were recorded with high Cl coal, despite the fact, that highest Cl content was found in NaCl impregnated coal.

HCl emission factor shows, that majority of the chlorine was released in the form of HCl regardless of the type of coal including coal impregnated with NaCl, fig. 3(a). When the NaCl impregnated coal was combusted, the HCl was released earlier in the combustion cycle, fig. 3(b). We assume, that NaCl during impregnation did not penetrate evenly through the coal grains but remained at elevated concentration on outer layers. However, the HCl concentration were higher during the whole combustion cycle when NaCl impregnated coal was used in comparison to low chlorine coal.

**Table 3. PCDD/F concentrations in flue gases and emission factors in relation to the Cl content in coals**

	Coal Cl content	PCDD/F concentration in flue gases	PCDD/F emission factor
	[%]	[ng TEQ/m <sup>3</sup> ]	[μgTEQ/t]
Low Cl coal	0.074	0.466	3.77
Coal mix	0.0979	1.19	9.02
Coal mix	0.1218	0.738	6.32
Coal mix	0.1696	2.39	23.3
Coal mix	0.2174	13.2	137
High Cl coal	0.313	105	1326
NaCl impregnated low Cl coal	0.378	34.4	271



**Figure 3. HCl emission factor in relation to the Cl content in coal (a) and time dependence of the HCl concentration in flue gases for combustion of different coals (b)**

The comparison of the PCDD/F emissions obtained from two coals and their mixtures does not provide complete evidence of the coal chlorine content influence on the PCDD/F emissions. Inhibitors of the PCDD/F formation were identified, like for instance sulphur and nitrogen compounds [16]. Low chlorine coal has higher sulphur content and higher SO<sub>2</sub> concentrations in the flue gases than high chlorine coal. It has to be noted, that lower temperatures of the flue gases in the chimney were recorded using low chlorine coal in comparison with high chlorine coal, due to difference in heating values. That could have negative impact on the formation of the PCDD/F in the chimney in the case of low chlorine coal.

Possible influences of different inhibitors or catalysers and different temperature profiles of the flue gases in the chimney on the PCDD/F can be excluded when comparing emissions from low chlorine coal and the same type of the coal yet impregnated with NaCl. The increase of PCDD/F emissions by two orders of magnitude due to coal impregnation with NaCl confirms decisive influence of the chlorine content in coal on PCDD/F emissions.

It was shown in the case of waste incinerators that the organisation of the combustion process and operation practice rather than the chlorine content of the input material influence the PCDD/F emissions [16]. The comparison of the simple stove for domestic heating and sophisticated waste incinerator is not adequate. From the viewpoint of the organisation of the com-

bustion process there are more similarities between coal combustion in residential stoves and open burning of waste in barrels. When burning waste in barrels it was also found that higher chlorine content leads to elevated PCDD/F emissions [17].

*Mass balance of the PCDD/F emissions via flue gases, chimney soot, and bottom ash*

PCDD/F were determined in the chimney soot and bottom ash when both low and high chlorine coal was used. The chimney soot from the experiments with insulated chimney and chimney with cooled heat exchanger was analysed. Only the bottom ash from combustion experiments with insulated chimney configuration was analysed, as it is assumed, that different chimney configurations do not influence the PCDD/F content in bottom ash.

The use of the high chlorine coal resulted in higher PCDD/F content not only in the flue gases, but also in bottom ash and chimney soot (tab. 4). The bottom ash contained low concentrations of PCDD/F. In the case of low chlorine coal some congeners were even below the limit of quantification. The chimney soot contained higher concentrations of PCDD/F than the bottom ash. The chimney soot in the non-insulated chimney with heat exchanger installed displayed higher concentration of PCDD/F than in the insulated chimney. This could be explained by greater adsorption of the PCDD/F on the particulate matter on the chimney walls at lower temperatures.

**Table 4. Flue gases, chimney soot, and bottom ash PCDD/F emission factors for selected combination of coals and chimney configurations**

		High chlorine coal		Low chlorine coal	
		Insulated chimney	Cooled heat exchanger	Insulated chimney	Cooled heat exchanger
Flue gases PCDD/F emission factor	µg TEQ /t coal	1326	112	3.7	1.3
Chimney soot PCDD/F emission factor		11.6	21.5	0.64	1.7
Bottom ash PCDD/F emission factor		1.6	1.6	0.05	0.05
Total PCDD/F emission factor		1339	135	4.0	2.8
Flue gases PCDD/F emissions share	%	99.0	82.8	94.5	47.5
Chimney soot PCDD/F emissions share		0.9	16.0	4.2	50.6
Bottom ash PCDD/F emissions share		0.1	1.2	1.3	1.9

In general the majority of PCDD/F were emitted with flue gases, in particular at elevated PCDD/F emissions, where 99% of the total PCDD/F were released via flue gases pathway. The share of the flue gases emitted PCDD/F was lower when using low chlorine coal. In this case also the absolute value of the chimney soot emission factor is low.

### Conclusions

According to the performed investigations, it is possible to conclude that higher emissions of the PCDD/F during coal combustion in the stove for residential heating were measured,



than they have been recorded by other research groups. It has to be emphasized, that our experiments were performed with commercially available coal intended for residential use. Our results show that coal combustion in the residential stoves can result in extreme concentrations of the PCDD/F in flue gases, three orders of magnitude higher than in modern waste incineration plants.

The emissions of PCDD/F in the flue gases increase by two orders of magnitude when the chlorine content in the coal was increased from 0.07% to 0.38% by NaCl impregnation. This is the first published demonstration of the fundamental role of chlorine content in fuel in relation to the elevated PCDD/F emissions released from coal combustion in residential stoves.

The global average chlorine content is estimated to be 0.034% in hard coals and 0.012% in brown coals. Significant regional differences occur. In some coal seams in the United States of America, Poland, Germany, Russia, Ukraine, and Japan so called salty coals, with chlorine content even in excess of 1% could be found [18]. To limit PCDD/F emissions the use of high chlorine coal in the residential stoves must be avoided.

PCDD/F could be formed in the chimney when high and low chlorine coal is combusted. It is estimated that 90% of the PCDD/F in the flue gases are formed in the chimney in the case of high chlorine coal and insulated chimney when combusting low chlorine coal formation in the chimney is less pronounced. Formation in the chimney should be taken into account when interpreting the PCDD/F emission measurements from coal combustion in stoves. Thermal properties of the chimneys used in residential dwellings and in emission measurement facilities usually differ. This is of particular importance when measurement results are used to derive emission factors for PCDD/F emission inventories.

The obtained results with heat exchanger installed in-between the stove outlet and chimney duct do not show marked decrease of the PCDD/F emissions. However, we consider, that the optimisation of the installation and the construction of the heat exchanger offer the potential to decrease the PCDD/F emissions. Heat exchanger additionally increase the thermal efficiency of the heating appliance, thus having a potential for a low cost win-win PCDD/F abatement measure.

Flue gases are the main PCDD/F emissions pathway from coal combustion in stoves. Lower amounts PCDD/F are found in chimney soot and bottom ash. However chimney soot in particular has to be appropriately deposited to prevent the contamination of the environment with PCDD/F.

Although it was projected that wood and coal use in residential heating appliances in urban areas will decline throughout the Europe, this trend stopped or even reversed in some areas. This is partially caused by high energy prices, which induced low-income households in particular to opt for the cheaper alternatives [19]. That is why the PCDD/F emissions from coal combustion in the residential stoves will continue to pose an environmental issue in the future, if not approached appropriately.

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