

High emissions of PCDD/Fs from coal fired stoves: indications of the formation in the chimney

Bostjan Paradiz¹, Jiří Horák¹, Panagiota Dilara¹, Gunther Umlauf¹

¹Institute for Environment and Sustainability, Joint Research Centre of the European Commission

Introduction

Emissions from solid fuels fired heating appliances in residential sector are considered to have a significant share in total PCDD/F emissions into the air, especially in some countries, where such appliances are widely used. In the year 2000 the dioxin emission inventory for the European Union attributed 21% of the total PCDD/F emissions to the residential combustion of wood and 8% to the residential combustion of coal¹. However, the emission factors for residential combustion of coal and wood are highly uncertain. Very high emissions of PCDD/Fs up to 660 µg TEQ/ton for coal combustion in stoves were reported for the first time by the Austrian Federal Environment Agency when sampling emissions from combustion of hard coal of Polish origin². The experiments latter performed by the North Rhine-Westphalia State Environment Agency gave values up to 133 µg TEQ/ton for coal combustion in stoves¹.

The conditions and mechanisms that could lead to high emissions from residential coal combustion are not yet entirely understood. Besides the uncertainty in the PCDD/Fs emission inventories this also prevents the conception and implementation of abatement measures. The gap in knowledge was already identified within the "Community Strategy for Dioxins, Furans and PCBs", which has attributed the highest research priority to the domestic combustion of wood and coal.³

The conventional coal fired stoves have very low energy efficiency due to the incomplete combustion and heat losses through the chimney⁴.

Temperatures of the flue gases at the stove exit in excess of 500° C, together with residence times of some seconds in the chimney at temperatures above 300° C suggests PCDD/F formation. Therefore an experimental program was prepared to investigate the effects of the temperature profile in the chimney on PCDD/Fs emissions. The experimental programme was designed to address also the possible memory effects when changing the operational parameters.

Materials and methods

A commercial low-cost stove of Polish origin, having advanced construction with down-draught combustion, was used for the experiments. After the optimisation of combustion parameters (primary air, flap controlled natural draft) the emissions of CO, particulate matter (PM) and volatile organic compounds (VOC) were low in comparison with the default emission factors for coal fired stoves.⁴ A stainless steel duct with the diameter of 136 mm and the height of 12 m served as chimney. Two PCDD/Fs sampling points were used, the bottom one at the distance of 0.6 m and the top one at 6.3 m from the stove outlet to the chimney. Different temperature profiles in the chimney were obtained by insulating the whole length of the chimney with a 25 mm thick mineral glass insulation. A commercial Polish hard coal having calorific value 30.0 MJ/kg was used for combustion experiments. The chlorine and sulphur content of the coal were 0.31 % and 0.32%, respectively.

Each combustion cycle consisted of two phases. During the 1 h initial phase hot ash was prepared. It served for the ignition of the main batch of 5 kg coal in the operational phase. During the operational phase the complete main batch of the coal was combusted - its duration was determined to obtain the same amount of the hot ash on the stove grating as there was at the beginning. The operational phase lasted for 3h 45min. The PCDD/Fs sampling start coincided with the addition of the main batch of coal and was performed during the whole duration of the operational phase of the combustion cycle.

The PCDD/Fs sampling was performed according to the standard EN 1948 – cooled probe method. Due to the low velocity of flue gases in the chimney the direct measurements by the Prandtl-Pitot tube were 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 carbon content and consumption of the fuel as well as the composition of the flue gases. The volume of approximately 1 Nm³ was taken during each sampling. Extraction, clean up and analysis of PCDD/Fs was performed in line with the EN 1948 standard. An aliquot of the extract was used also for PAH analysis. The CO, CO₂, NO, SO₂ and HCl concentrations in the flue gases were measured by FTIR and VOC by FID.

During the first series of the combustion tests the chimney was not insulated. PCDD/Fs samplings were performed at the top sampling point during the 2nd, 5th and 8th combustion test. The second series of combustion tests, numbered from 9 to 16 was performed with the insulated chimney. PCDD/Fs samplings were performed at the top sampling point during the 10th, 12th and 16th test, while during the 13th and 15th test the bottom sampling point on the chimney was used.

Results

Very high emissions of the PCDD/Fs were observed in all combustion experiments (Figure 1). Concentrations of PCDD/Fs in flue gases were between 6 and 115 ng I-TEQ/Nm³ recalculated to 11 % of O₂. These values are comparable to those of waste incinerators with minimal air pollution control⁵.

No significant changes in PCDD/Fs concentrations were observed in subsequent measurements with the same chimney configuration (tests 2, 5 and 8; tests 10, 12 and 16, tests 12 and 14). The memory effect in this experimental set-up is therefore not pronounced. Therefore, one preconditioning combustion cycle is deemed to be enough to prevent possible influences of the memory effect on the PCDD/Fs emission measurements when changing the experimental configuration.

Emissions of PCDD/Fs were particularly high when the chimney was insulated – they were one order of magnitude higher than those obtained with the non-insulated chimney. At the same time, the temperatures of the flue gases in the insulated chimney were approximately 100° C higher than in the un-insulated chimney (Table 1). The emissions of other pollutants including PAHs were not affected by the different temperature profile in the chimney. The temperature in the chimney did therefore not influence the combustion processes in the stove. These results provide significant evidence that the reactions in the chimney at higher temperatures can contribute to very high emissions of PCDD/Fs.

Figure 1. PCDD/Fs concentrations in flue gases.

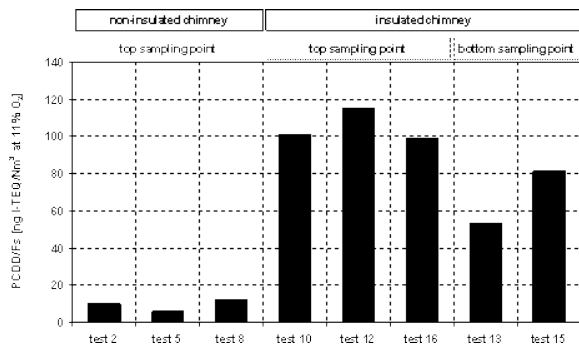


Table 1. Average emission factors and temperatures of flue gases in the chimney

Parameter	unit	non-insulated chimney	insulated chimney	
		top sampling point (3 samples)	top sampling point (3 samples)	bottom sampling point (2 samples)
PCDD/F (I-TEQ)	µg/t	126	1326	820
Sum PAH (16-EPA)	g/t	63	55	64
Benzo[a]pyrene	g/t	1.3	1.5	1.7
CO	kg/t	14.2	16.3	13.1
NO _x (as NO ₂)	kg/t	3.6	3.4	3.6
SO ₂	kg/t	3.2	3.0	2.6
VOC	kg/t	4.8	9.3	6.1
PM	kg/t	3.1	3.8	4.1
HCl	kg/t	2.7	2.6	2.5
T1 – d=0.6 m ***	°C	332	419	397
T2 – d=1.8 m*	°C	260	385	367
T3 – d=3.0 m*	°C	228	353	333
T4 – d=4.1 m*	°C	193	317	302
T5 – d=5.3 m*	°C	166	288	277
T6 – d=6.3 m****	°C	146	264	252

*distance from the stove flue gases exit **bottom PCDD/Fs sampling point *** top PCDD/Fs sampling point

The average residence time of the flue gases in the chimney is estimated to be approximately 0.5 and 5 seconds when the sampling was performed at the bottom and top sampling point respectively. The concentration of the PCDD/Fs at the bottom sampling point of insulated chimney was an order of magnitude higher than PCDD/Fs concentration at the top sampling point of the non-insulated chimney. On the other hand the concentrations of the PCDD/Fs in the insulated chimney at the bottom sampling point were only approximately one third lower than at top sampling point. This implies that the higher temperature of the flue gases has a more pronounced effect on the dioxin formation than the residence time.

Emission measurements of the small combustion appliances are often performed in laboratory conditions with a chimney, which does not have the same thermal capacity nor conductivity as the chimneys used in residential dwellings. If PCDD/Fs measurements are intended to provide emission factors to be used for emission inventory compilation, particular attention should be given to use the chimneys and stove connections to it that are representative for residential dwellings. In this respect PCDD/Fs samplings performed in the chimneys of residential dwellings have a clear advantage.

Experiments presented in this paper were not aimed to provide emission factors for coal combustion in the stoves. However the results obtained support the inclusion of the new emission factor 400 µg TEQ per ton for high chlorine coal combustion in the residential stoves in the new version of the Standardized Toolkit for Identification and Quantification of Dioxin and Furan Releases⁵.

Acknowledgement

The authors would like to thank Ms. Krystyna Kubica, Nilu Polska for her assistance in arranging for the stove and the coal as well as for her overseeing the construction and operation of the experimental combustion facility. We would like to acknowledge Dominique Lesueur, Andrea Brunella and Denis Fachinetti for constructing the combustion facility. In addition Denis Fachinetti is further acknowledged for his assistance in performing the combustion experiments.

References

1. Quass U., Fermann M. and Bröker G. (2000) The European Dioxin Emission Inventory, Stage II, North Rhine-Westphalia State Environment Agency, Germany http://europa.eu.int/comm/environment/dioxin/pdf/stage2/volume_1.pdf
2. Moche W. and Thanner G. (1998) *Organohalogen Compounds* 50: 329-332
3. Community Strategy for Dioxins, Furans and Polychlorinated Biphenyls, Communication from the Commission to the Council, the European Parliament and the Economic and Social Committee COM(2001) 593 final
4. Kubica K., Paradiz B., Dilara P., Klimont Z., Kakareka S., Debski B. (2004) Small combustion installations in Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, Third Edition, Copenhagen: European Environment Agency
5. UNEP Chemicals (2005), Standardized Toolkit for Identification and Quantification of Dioxin and Furan Releases, 2nd edition, Geneva, Switzerland