PCDD/F AND PAH EMISSIONS FROM DOMESTIC HEATING APPLIANCES WITH SOLID FUEL

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Introduction

The use of domestic heating appliances with solid fuel is suspected as an important source of polychlorinated dibenzodioxins and polychlorinated furans (PCDD/F) in Flanders. Because of the broad range of concentrations reported in the literature[1-4] and the lack of data for typical appliances and fuels used, several types of wood and coal furnaces with different fuel compositions were tested. The aim of the work was to develop more refined emission factors for the various applications.

Methods and materials

Five different heating systems were selected, three wood stoves and two "multiburners", suitable for coal, wood and lignite. Table 1 gives an overview of their most important characteristics.

Table 1: Characteristics of the selected heating appliances according to the manual

Type	Fuel	Power, kW	$Ø_{out}$, mm	Weight, kg	BxHxD, mm
Dovre Multi 750 GM	Wood, lignite	9-11	150	190	700x705x490
Dovre Multi 759 GH	Wood, lignite, coal	9-11	150	190	700x705x490
Flam Kameleon 44/70	Wood, lignite, coal	7	200	245	615x886x530
Cardoen CA4	Wood	8	150	-	650x980x440
Jøtul 118	Wood	2-14	125	117	

The stoves were connected to a vertical, insulated stainless steel chimney with an internal diameter of 25 cm and a length of 3 m. The flue gases were diluted with ambient air by a funnel system with adjustable gap and two fans. The duct for the diluted flue gases had an internal diameter of 18 cm and a mixing section of 2 m. The vertical measuring duct with a total vertical length of 4 m was equipped with sufficient measuring holes for the various sampling systems. The burning rate was measured by placing the stove on a scale that registered the weight continuously. A scheme of the installation is presented in Figure 1.

The stoves were operated as commonly done by first producing a glowing fire with a small amount of fuel and then filling to obtain a nominal heat output. The whole burning cycle, from start till stop, took 6 hours. During this period flow, temperature and the concentrations of CO, CO_2, NO_x, SO_2, O_2 and TOC were measured continuously. Dust, PAHs and PCDD/F were sampled for 1, 2 and 6 hours respectively. For PAHs and PCDD/F, the filter-condensation method was used with Tenax as solid sorbent. PAH (16 EPA congeners) samples were analyzed by LRGC/MS after

A(ccelerated) S(olvent) E(xtraction) and PCDD/F samples by HRGC/HRMS according to EN 1948.
Tested fuels were different types of coal and wood eventually mixed with household waste. From each fuel composition a representative sample was collected and analyzed for N, C, S, Cl and caloric value.

Figure 1: Experimental set-up

According to EN 13240 "Roomheaters fired by solid fuel-requirements and test methods" the results are referred to 13% O_2. Because of the high dilution rate in the funnel sampling system the recalculation was performed on a CO_2 base as in coal and wood fire 13% O_2 corresponds with 8% CO_2.

Results and Discussion

Coal appliances

The coal appliances were tested 6 times: 2 times with anthracite at nominal heat output and 3 times at reduced output, once lignite was used at nominal output. In all cases the gas flow was maintained at about 800 Nm^3/h. While at nominal heat output the dioxin sampling enclosed the whole burning cycle, at reduced output the ignition, normal working and burn out periods were sampled separately on 3 different days and the combined samples of each period were analyzed. The results are summarized in table 1.

Table 1: Concentrations of PAHs and dioxins in the emissions of coal stoves

Fuel	Heat output Appliance(*)	PAHs mg/Nm^3	PCDD/F ng I-TEQ/Nm^3	
Anthracite	Nominal - 1	2.1	0.13	
Anthracite	Nominal - 2	3.57	25.5	
Lignite	Nominal - 1	4.2	1.5	
Anthracite	Reduced - 1	12.9	Ignition	0.748
		13.9	Normal	0.360
		21.9	Burn out	0.108

(*): 1= Dovre Multi 750 GM; 2= Flam Kameleon 44/77

The concentrations of PAHs and PCDD/F during anthracite and lignite burning are rather low, less than 22 mg/Nm^3 for PAHs and, with one exception, less than 2 ng TEQ/Nm^3 for PCDD/F. Dioxins are emitted primarily during ignition, while PAHs are more abundant during burn out.

Wood stoves

In total 19 experiments were performed with woodstoves burning pure pine and hardwood of different ages and sizes as well as treated wood and mixtures of wood with the combustible fraction of household waste. The average gas flow in the measuring duct was situated between 350 and 400 Nm^3/h. Similar to the coal fired furnaces some tests were performed at a reduced heat output. The results of the corresponding PAH and PCDD/F analyses are summarized in Table 2.

Table 2: Concentrations of PAHs and dioxins in the emissions of coal stoves

Fuel	Heat output Appliance (*)	PAHs mg/Nm^3	PCDD/F ng I-TEQ/Nm^3
Hardwood, > 2 year	Nominal – 1	5.84	2.69
Hardwood, > 2 year, middle sized	Nominal – 1	5.98	7.26
Hardwood, > 2 year, small sized	Nominal – 1	6.45	2.71
Hardwood, < 1 year	Nominal – 1	6.03	2.91
Pinewood, < 1 year	Nominal – 1	4.17	2.60
Pine- and hardwood, < 1 year	Nominal – 1	2.86	0.4
Pine- and hardwood, < 1 year	Nominal – 2	53	0.26
Pine- and hardwood, < 1 year	Nominal – 2	48.4	0.64
Pine- and hardwood, < 1 year	Nominal – 3	33.2	0.58
Pine- and hardwood, < 1 year	Nominal – 4	13.8	0.73
Treated wood	Nominal – 1	7.73	200
Co-combustion of household waste	Nominal – 1	4.03	10.8
Co-combustion of household waste	Nominal – 2	116	172
Co-combustion of household waste	Nominal – 2	57	5.88
Co-combustion of household waste	Nominal – 3	15.6	20.4
Co-combustion of household waste	Nominal – 4	13.1	0.69
Pine- and hardwood, < 1 year	Reduced – 2	56.2	14.1
Pine- and hardwood, < 1 year	Reduced – 3	10.8	1.0
Pine- and hardwood, < 1 year	Nominal – 1	8.3 8.3 7.5	Ignition 0.66 Nominal 0.81 Burn out 2.48

(*): 1 = Dovre Multi 750GH; 2 = Flam Kameleon 44/70; 3 = Cardoen C4A; 4 = Jøtul 118

Increased PCDD/F emissions during the combustion of clean wood under conditions of nominal heat output were observed for stove type 1. Untreated wood had the lowest PAH and dioxin emissions. Mixed young wood generally had lower PCDD/F but higher PAH emissions than well dried hardwood. Reduced heat output resulted in increased PCDD/F concentrations. Ignition and burn out periods or the age of wood do not show large differences in the emission concentrations. Only when household waste or treated wood is burned, the PCDD/F concentrations increase significantly.

Determination of emission factors

From the concentrations in the flue gas, emission factors were calculated by multiplying the concentration, the flow and the sampling time of the individual experiments, and dividing the result by the amount of fuel consumed. The results for PCDD/F and PAHs are summarized in Table 3.

An evaluation of the data shows a large spread, in some cases over several orders of magnitude.

Table 3: Survey of emission factors obtained from this study

Type of appliance	Emission factor for dioxins in ng I-TEQ/kg fuel			
	Mean	Median	Minimum	Maximum
Coal stoves with anthracite	77.1	4.3	1.15	282
Wood stoves with untreated wood	24.4	18	1.97	88.7
Wood stoves with wood and waste	350	106	3.85	1411
Wood stoves with treated wood	1702	1702		
Type of appliance	**Emission factor for PAHs (16 EPA) in mg/kg fuel**			
	Mean	Median	Minimum	Maximum
Coal stoves with anthracite	68.5	41	33.5	158
Wood stoves with untreated wood	149	60	24	428
Wood stoves with wood and waste	328	140	35.7	902
Wood stoves with treated wood	53.9	53.9		

Relationship between CO, PAHs and PCDD/F

According to EN 13240, roomheaters fired by solid fuel are divided in classes according to their CO emission levels. For that reason the possibility of the application of the CO-concentration as a criterion for the presence of PAHs and/or PCDD/F was checked. Figure 2 demonstrates the relationship between the three parameters. To a small degree, CO can be considered as an indicator for PAHs but no significant correlation is found between CO and PCDD/F emissions.

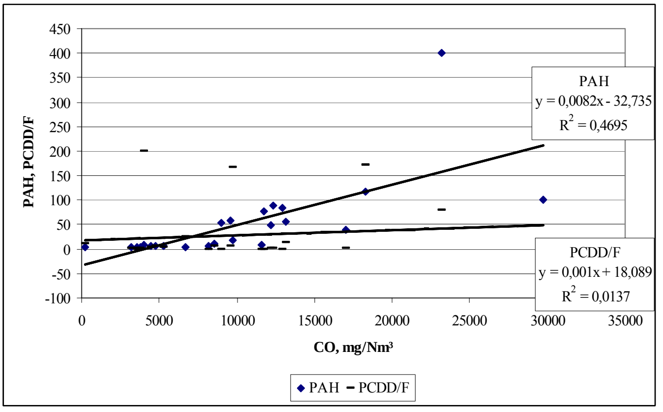

Figure 2: Relationship, CO, PAH, Dioxins

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