PCDD/F-Emissions from Coal Combustion in Small Residential Plants

Wolfgang Moche, Gerhard Thanner

Federal Environment Agency, Spittelauer Laende 5, 1090-Vienna, Austria

Introduction

According to the results of recent studies[1,2] in Austria PCDD/F-emissions are mainly produced by non-industrial combustion plants and production processes. The estimated emissions of this sector amount to 16 g I-TEQ /year representing 58% of the total dioxin emissions in Austria.
Most of the corresponding emission data presented in the international literature deal with wood combustion in various types of heating furnaces. The results of experiments[3,4], carried out at test benches, showed that the PCDD/F-emissions of household stoves with natural wood as fuel are in the range of 0.01 to 0.1 ng I-TEQ/Nm³ (11% O_2) . The co-combustion of woodwaste and household waste increases the PCDD/F-emissions significantly with maximum concentrations of 114 ng I-TEQ/Nm³ (11% O_2).
There are only few data available for PCDD/F-emissions from combustion of coal in small residential plants. The emission concentrations for different types of coal and coke as reported by Bröker et al. (1992)[3] are in the range of 0.015 to 0.14 ng I-TEQ/m³ (0% O_2).
Therefore the Federal Environment Agency started a programe to obtain further data on emissions of PCDD/F from combustion of coal in small residential plants under field operation conditions. This paper presents first results of the emission measurements.

Experimental

Furnaces: a) central heating boiler, 32kW (for coal/coke)
b) household stove, 6 kW

The stoves used have been in service for more than 10 years for household heating. They have been operated without any modification at their initial installation places.

Fuel: The coal used for the experiments was hard coal purchased from a local dealer.

Sampling: First experiments with a filter/cooler-sampling train according to VDI-3499 (part 2)[5] showed to be inappropriate due to the high amounts of coal dust in the fluegas resulting in rapid clogging of the filter-compartment. This made continuous sampling impossible. The finally used sampling train consisted of a water-cooled quartz probe with a titanium-nozzle attached, a condensation flask, two impingers each of them containing 750 ml ethyleneglycol at -10°C, a glasfibre-filter and three PU-foam plugs (Ø 4cm, h 5cm). The condensation flask was spiked with ^{13}C-1,2,3,4-TCDD as sampling spike. The sampling was done isokinetically with simultaneous monitoring of CO/CO_2- and O_2-concentration. The sampling site had been located approximately 8 m above the furnaces to obtain optimal conditions for isokinetic sampling. The sampling volumes comprised 4 to 6 m³ with sampling periods of 2 to 4 hours.

Analysis: The condensate and the ethyleneglycol were pressure filtrated over 0.45 µm teflon filters. The filtrated impinger/condensate-fractions, the combined teflon and glasfibre filters and the PU-foam plugs were spiked with a ^{13}C-recovery standard solution containing all 2,3,7,8-substituted PCDD/F-congeners and extracted with toluene separately. The clean-up of the separate extracts comprised a four-stage column chromatography. The analysis and quantitation was carried out by HRGC/HRMS.

Sampling and analysis as described above are in compliance with the recently released CEN-1948[6].

Results

The results of the emission measurements and the derived emission factors are presented in the table below.

Furnace	experiment	% O_2	ng I-TEQ/Nm³ (0% O_2)	ng I-TEQ/kg coal
household stove 6 kW				
	#1	17.1	13.78	108.46
	#2	16.8	46.43	380.43
	#3	16.2	87.18	663.90
	#4	16.6	57.33	470.01
	#5	16.5	48.25	370.07
	#6	16.2	23.36	185.29
central heating boiler 32 kW				
	#1	16.3	0.11	0.29
	#2	16.1	0.07	0.33

The PCDD/F-emission concentrations of the central heating boiler are comparable with the data known from the literature.
Much higher emissions were observed when burning the same type of hard coal in an ordinary household stove. The homologue profiles of the central heating furnace, shown in figure 1, are comparable to combustion profiles known from the literature[8]. The homologue profiles of the emissions of the household stove, shown in figure 2, show decreasing abundance of the homologues with increasing chlorination degree for the PCDFs as well as for the PCDDs.

According to the statistics published by ÖSTAT (1995)[7] there are still 120 500 flats in Austria which are using household stoves fueled by coal, coke or coal-briquettes respectively. The annual consumption of coal/coke/briquettes of this sector has been estimated at 303 000 t (including 57 890 t hard coal) for 1992. This would result in annual PCDD/F-emissions in the range of 6 to 38 g I-TEQ/year only for hard coal combustion in household stoves. This would significantly increase the latest emission-estimation for non-industrial combustion (16 g I-TEQ/year). However, further investigations are needed to update the emission factors for coal combustion in small residential plants.

Figure 1: Homolgue profile of PCDD/F-emissions from coal combustion in a central heating boiler; average of two experiments

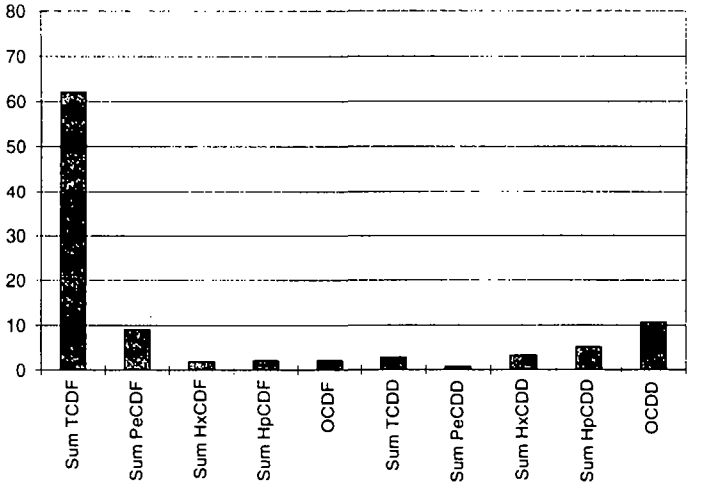
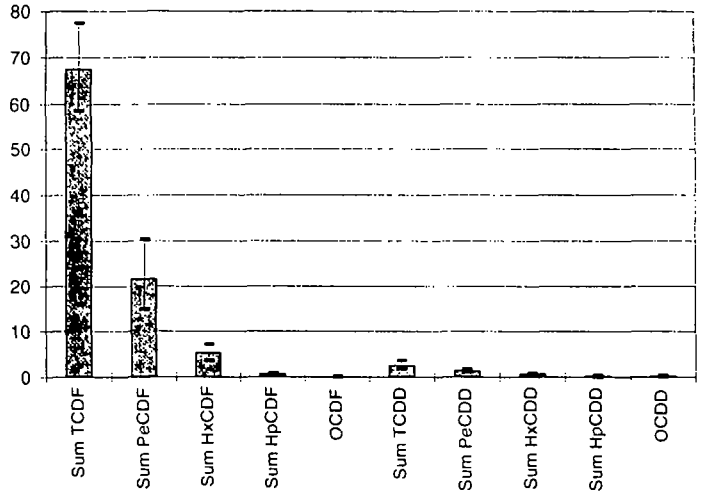


Figure 2: Homolgue profile of PCDD/F-emissions from coal combustion in a household stove; average, minimum and maximum of six experiments



References

1. König K, Radunsky K, Ritter M; Austrian Air Emission Inventory 1994; *Umweltbundesamt-Wien*, Reports R-140 **1997**.
2. Wurst F, Hübner C; Erhebung des PCDD/F-Emissionspotentials für Österreich; *FTU-Forschungsgemeinschaft Technischer Umweltschutz*, Wien **1997**.
3. Bröker G, Geuke KJ, Hiester E, Niesenhaus H; Emission polychlorierter Dibenzo-p-dioxine und -furane aus Hausbrand-Feuerungen; *LIS-Berichte* Nr. 103, **1992**.
4. Schatowitz B, Brandt G, Gafner F, Schlumpf E, Bühler R, Nussbaumer T; Dioxin Emissions from Wood Combustion; *Chemosphere* **1994**, Vol. 29, Nos 9-11, pp. 2005-2013.
5. Messen von polychlorierten Dibenzo-p-dioxinen und Dibenzofuranen; *VDI-Richtlinie* 3499 (Entwurf), Blatt 3, **1993**.
6. Stationary source emissions – Determination of the mass concentration of PCDD/PCDFs; *CEN* 1948, **1997**.
7. ÖSTAT; Energieverbrauch der Haushalte im Jahre 1992; *Beiträge zur österreichischen Statistik*, Heft Nr. 1.169, **1995**.
8. Rappe C; Dioxin, patterns and source identification; *Fresenius J. Anal. Chem.* **1994**, Vol. 348, Nos 1-2, pp. 63-75.