

# FORMATION AND SOURCES - POSTERS

## THE *DG ENV* EUROPEAN DIOXIN EMISSION INVENTORY - STAGE II: Elevated Emissions of Dioxins and Furans from Domestic Single Stove Coal Combustion

K.-J. Geueke, A. Gessner, E. Hiester, U. Quaß, G. Bröker

LUA NRW; PO Box 10 23 63; D-45023 Essen, Germany

### Introduction

Domestic combustion of fossil fuels used for room heating and/or cooking has frequently been discussed as one possible source of dioxin and furan emissions. During the last decade a number of investigations have been carried out focusing on different aspects of this topic. From various studies it can be concluded that combustion of liquid or gaseous fuels in common heating appliances do not lead to significant PCDD/F formation<sup>1, 2</sup>. By contrast, burning of solid fuels like coal, lignite and wood may cause increased dioxin emissions<sup>3</sup>. Of these fuels only wood combustion, in some European countries the predominant household energysource, was suspected to be a considerable dioxin emission source<sup>4</sup>. This is particularly due to the high emission factors found when contaminated wood (i .e. treated with wood preservatives like PCP or wood containing other chlorinated compounds) is combusted. As the amount and composition of contaminated wood burned in the households is not well known the dioxin emission can only be estimated with high uncertainty. By contrast, whenever investigated dioxin emissions from coal combustion were revealed to be comparatively low and a default emission factor of 2 µg TEQ per tonne fuel was applied for the emission estimation in the report on Stage I of the DG XI European Dioxin Project<sup>4</sup>. Using this factor, the overall annual PCDD/F release from domestic coal combustion in 17 European countries revealed to be only around 40 g I-TEQ (less than 1 % of total PCDD/F emissions).

However, in 1998 the Austrian Umweltbundesamt presented an unexpected high emission factor for combustion of hard coal from Poland in a simple constructed single heating stove<sup>5</sup>. Application of this emission factor (range: 108 to 663 µg I-TEQ/tonne fuel at 0% O<sub>2</sub>) would at least double the emission estimate for domestic combustion in Austria and would increase the Stage I European estimate by two orders of magnitude thus making domestic coal combustion the most relevant emission source in Europe.

LUA therefore decided to carry out a comparative measurement program in order to clarify whether the special fuel used in Austria or the stove construction or the combination of both may have caused the elevated emissions.

### Materials and methods

The measurements were carried out at a test rig for heating facilities. The stacks used are constructed similar to those commonly found in living houses (height: ca. 10 m, bricks, rectangular shape, cross section 15\*15 cm). Two stoves were tested which were taken from the

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collection of the Chimney Sweeper school. The first (stove A), being the elder one, operates as through-burning stove and has a power of ca. 4 kW. It is equipped with a primary air supply only and was produced between the years 1955 and 1962. The second stove (Stove B) is a more modern type that has been on the market about 20 years ago (produced 1983). It is a 6 kW under-burning stove with thermostat and secondary air supply. The ovens were connected to the stacks by stainless steel ducts about 1.5 m above ground level. The stoves were operated as commonly done by first producing a basic glowing fire through ignition of a small amount of fuel and then filling the main fuel loading onto the glowing bed.

PCDD/F samples were taken at 10 m above ground using the filter/cooler method according to VDI 3499 slightly modified by insertion of a quartz wool cartridge in front of the cooler. Sampling was not isocinetically due to the very low flue gas velocity. The sampling rate was ca. 2 m<sup>3</sup>/h with a 15 mm nozzle. Sampling started after the main fuel batch had reached a steady state burning. Air supply was regulated manually in order to maintain a temperature of about 250-300 °C in the fuel gas at the exit of the stove.

Six different fuels were applied. Besides the hard coal from Poland which was kindly provided by the Austrian Umweltbundesamt hard coal and lignite of German provenience (anthracite, hard coal briquettes, coke, lignite from Rhine area) and lignite from the Czech Republic was used.

### Results and discussion

In the following the results obtained for only four of the six fuels are presented. Analyses of the missing fuels are currently under way and will be included in the symposium presentation. Even without the missing results the main questions can be answered already.

Table 1 shows the concentrations of PCDDs and PCDFs found in the experiments. From fuel consumption determined by weighing the fuel input and the amount of residue materials after completion of the measurement fuel-related emission factors can be derived which are shown in table 2.

Surprisingly, the more modern stove B generally caused higher emissions than the more simple stove A. A possible explanation for this effect could be seen in the different condition of operation. According to the under-burning principle in Stove B the fuel is not pre-heated by the flue gases like it is the case in the through-burning stove. Hence, in Stove B cold, not yet degassed fuel enters the burning zone. This leads to a worse burn-out (indicated by higher CO concentrations) and to higher content of volatile hydrocarbons in the flue gases which might produce higher PCDD/F concentrations. By contrast, in the more simple through-burning stove A the entire fuel batch is pre-heated and degassed during the first minutes of burning thus leading to lower smouldering gas production at steady state conditions.

Compared to results obtained in previous studies all flue gas concentrations observed appear to be elevated. For instance, measurements carried out in 1992 by our own agency<sup>3</sup> using a 32 kW central heating unit for solid fuels did not yield concentrations above 0.1 ng I-TEQ/m<sup>3</sup> (referred to 0% O<sub>2</sub>) when any type of coal was combusted. Further, experiments made by Rheinbraun on behalf of The German Umweltbundesamt with a modern through-burning single stove (7 kW) also

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revealed low emissions (29 to 234 pg TEQ/m<sup>3</sup> referred to 0 % O<sub>2</sub>); this observation also supports the above mentioned assumption that through-burning stoves have lower emissions during steady state operation. As the stove used by Rheinbraun was a more modern one than stove A used in our experiments, a better burn-out could be achieved which coincide with lower oxygen content in the flue gas during operation (about 13% compared to ~18% in our experiments).

Apparently, the Polish hard coal leads to significantly elevated PCDD/F emissions compared to the other fuels. This trend therefore is in agreement with the observation made by the Austrian UBA. These observations thus can be considered as principally being plausible. However, even the highest concentration found in our tests did not reach the lowest value observed in the Austrian UBA experiments. In view of the variation of PCDD/F concentration with stove operation conditions observed in the tests presented here it appears possible even worse combustion conditions might have caused the Austrian measurement results.

## Acknowledgements

The authors are greatly indebted to the North Rhine-Westphalia Chimney Sweeper Association for providing the stoves and the test stacks and for their excellent support before and during the measurement campaign. Thanks also to W. Moche of the Austrian Umweltbundesamt for his help to purchase the Polish hard coal. Financial contribution by the European Commission, DG Environment is gratefully acknowledged.

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pg I-TEQ/m <sup>3</sup> at 0% O <sub>2</sub>	Stove A				Stove B			
	Sample 1	Sample 2	Sample 3	mean	Sample 1	Sample 2	Sample 3	mean
Lignite Germany	197	405	396	333	796	493	350	546
Lignite Czech Rep.	292	30	20	114	172	199	229	200
Anthracite	655	352	547	518	1842	1178	884	1301
Hard coal Poland	2625	4494	2207	3109	6774	8098	15642	10171
Comparison with Austrian values:	13780 - 87180 pg I-TEQ/m <sup>3</sup>							

**table 1 Measured PCDD/PCDF concentrations (corrected for oxygen content)**

EF (µg I-TEQ/ton fuel)	Stove A				Stove B			
	Sample 1	Sample 2	Sample 3	mean	Sample 1	Sample 2	Sample 3	mean
Lignite Germany	0,80	1,83	2,40	1,68	1,84	1,20	1,04	1,40
Lignite Czech Rep.	1,26	0,11	0,06	0,48	0,48	0,54	0,55	0,52
Anthracite	3,15	1,77	1,95	2,29	8,19	4,40	0,03	4,21
Hard coal Poland	14,59	19,91	11,08	15,19	21,84	25,87	55,24	34,31
Comparison with Austrian values:	108,5 - 663,9 µg I-TEQ/tonne							

**table 2 Emission factors derived from measured concentrations and fuel consumption.**