FORM

EMISSION OF DIOXINS FROM DANISH WOOD-STOVES

Vikelsøe, J.[A], **Madsen, H.**[B] **and Hansen, K.**[C]
[A] National Environmental Research Institute, P.O.Box 358, 4000 Roskilde, Denmark
[B] Danish Prognosis Information, Hvedevænget 38, 2980 Kokkedal, Denmark
[C] Danish Technological Institute, Teknologiparken, 4000 Århus C, Denmark

Introduction

These studies, initiated and funded by the Danish government, represent the final investigations in the Danish Incinerator Dioxin Program. In a preliminary study of different wood-stoves and fuels[1], very high emission of dioxins were found, implying that the annual Danish emission of dioxins from this source would be of the same order of magnitude as the emission from the Danish incinerators. A follow-up study was subsequently carried out, involving new experiments at another test site at the Danish Technological Institute (DTI), as well as method development and validation studies at the National Environmental Research Institute (NERI)[2]. However, the follow-up study failed to confirm the previously found high emissions. The data indicated that the high values found in the preliminary study could have been caused by carry-over occurring in the chimney of the test site, the dioxin being formed in experiments burning PCP-impregnated wood included in that study. The present investigation takes this problem into consideration and makes use of improved testing, sampling, and analytical methods.

Objectives

To estimate the annual emission of dioxins from wood-stoves in Denmark, to make a decision ground for reducing the dioxin emission from this source as far as possible, and to make a basis for instructions for users and manufacturers of wood-stoves.

Experimental plan

4 types of stoves, 3 types of fuels (all pure wood), and 2 operating conditions were tested.

Types of stoves: Type L and type M (both included in the previous studies[1]), type Ln (type L new model) and Type P (experimental 2-stage combustion prototype designed by DTI, using preheating of air and inverted combustion, i.e. downward flame direction in main combustion chamber). The stoves were designed for additional heating of single rooms, the heat-output ranging from 3.5 kW to 9.2 kW.

Types of fuels: Beech (reference fuel in the previous study[1]), birch (Danish reference fuel) and spruce (fuel commonly used in Denmark).

Operating conditions: Normal operation (as expected for non-experts) and optimal operation (well controlled operation with good combustion, i.e. CO-emission as low as possible).

An experimental plan was designed, comprising all combinations of stoves, fuels and operating conditions, making a total of 24 tests, carried out in a pre-randomized sequence. The dioxin samples were analyzed blind in a second pre-randomized sequence, i.e. the identity of the individual samples was unknown to the analysts.

Methods

The wood-stove experiments were performed at DTI at a special test facility. Two experiments were performed a day, each comprising a full combustion cycle of at least 3 hours duration. Carry-over from old deposits was avoided by the use of a new system, and by omitting the burning of PCP-containing wood. The wood was harvested in Danish forests near the test site, cleaved, and equilibrated to about 18 % absolute moisture. Between 4 and 8 kg of fuel, depending on the size of the stove, was burned in each experiment. In the experimental setup, the stove was connected to a chimney followed by a dilution tunnel system, which diluted the total stream of flue gas about 1 to 10. A data logging system registered CO_2, CO, temperatures in chimney and tunnel, hydrocarbons, HCl, draught and flow.

The dioxin samples were taken by DTI in the dilution tunnel isokinetically according to the Nordic recommended method[3]. A volume of about 1 m^3 was sampled during a whole combustion cycle.

The dioxin analysis were performed by NERI. The samples were extracted by refluxing 24 hours in toluene using water-removing equipment[4], the clean-up performed on 30 g of silica and 4 g of basic alumina. Carbon clean-up was omitted. The samples were diluted 1 to 4 before GC/MS, which was performed on a SE-54 column as group-specific analysis. Laboratory spikes were used for recovery correction for congener groups individually, the spike mix containing 1 carbon-labelled isomer in each congener group.

Validation of the tunnel-sampling in combination with the modified analytical method was done by comparison against the method used in the preliminary study[2]. Stored as well as new samples were analyzed with both methods. No statistical significant difference between levels were found, but the new method had significantly lower standard deviation. Some of the samples were also analyzed by RIVM, Bilthoven, the Netherlands, with results on the same level as the values found by NERI.

Results, statistics and discussion

The main emission from all experiments was TCDF and some TCDD. The results from all experiments are shown in Fig. 1 as total PCDD + PCDF in ng/Nm^3 flue gas.

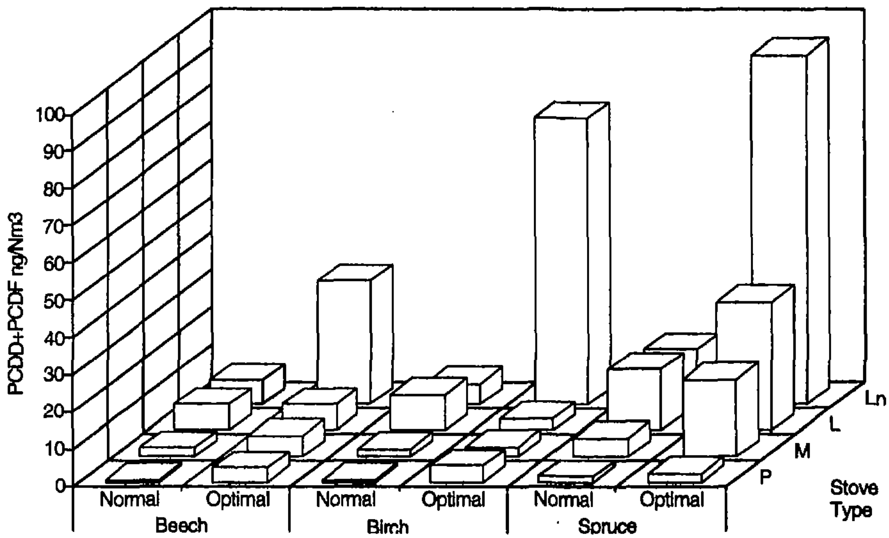

Fig. 1 Experimental Results. Dioxin emission from Danish wood-stoves.

A statistical analysis was performed by the Danish Prognosis Information (DPI), based on general linear models. The following statistical model involving active (direct controllable) parameters was found:

$$\log(\mathrm{DIOXIN}) = \mu(\mathrm{STOVE}) + \mu(\mathrm{FUEL}) + \mu(\mathrm{OPERATION(STOVE)}) + \varepsilon$$

That is, the dioxin emission depends upon type of stove and on the type of fuel, but the influence of operation depends upon the type of stove. ε is combined model and analytical error.

- The statistical analysis show highly significant differences between stoves, between fuels, and between optimal and normal operation for each stove.
- The standard error in the present study is only the half of the error in the preliminary study.
- There is no interference between STOVE and FUEL, i.e. the influence of FUEL is the same for all combinations of stove and operation.

- The P-stove has significantly lower emission than the others, and the M-stove has lower emission than the L- and the Ln-stove.
- The emissions from birch and beech do not have any significant difference, but are significantly lower than the emission from spruce (average 42 % of spruce).
- The "optimal" operation leads, somewhat surprising, to significant higher emission for the Ln-stove and the P-stove (average 230 % of normal), but not for the other stoves.

The statistical analysis was extended to include passive parameters, such as temperatures, draught, heat-output, CO, HCl etc. Surprisingly, the introduction of passive parameters did not significantly improve the model fit. However, if all active parameters were neglected, a highly significant positive correlation between emission and heat-output was found.

Annual emission

To estimate the annual emission, the statistical analysis was repeated using the dioxin emission per kg of fuel. Again a model was found, which compared well with the flue gas concentration model. However, the influence of fuel was more pronounced, the birch and beech emitting on the average 34 % of spruce. The tox-equivalents were estimated assuming the same isomer distribution in each group as found for MSW-incineration.

The annual emission from Danish wood-stoves was then computed, using the estimated numbers of stoves, and the amounts of the various fuels sold, to 0.40 g Nordic tox-equivalents / year. This is roughly 100 times lower than the annual emission from incinerators estimated in the Danish incinerator study[5].

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