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EFFECT OF NON-STATIONARY COMBUSTION PHASES ON EMISSION FACTORS OF SELECTED POLLUTANTS AND PCDD/F FROM DOMESTIC COMBUSTION

VLIV NESTABILNÍCH SPALOVACÍCH PROCESŮ NA EMISNÍ FAKTORY VYBRANÝCH ZNEČIŠŤUJÍCÍCH LÁTEK A PCDD/F Z MALÝCH ZDROJŮ

Abstract

In four boilers was combusted bituminious coal and lignite. In flue gases there were determined these pollutants: PM, TOC, CO, PAHs, PCBs, PCDD/Fs. It was assessed the effect of stoking period, wet fuel and lower output on emission factors of above mentioned pollutants. It was find out that values of emission factors were significantly higher than emission factors obtained during tests under normal combustion conditions.

Abstrakt

Ve čtyřech typech kotlů malých výkonů bylo spalováno černé a hnědé uhlí. Ve spalinách byly sledovány znečišťující látky (PM, TOC, CO, PAU, PCB, PCDD/F). Byl sledován vliv přikládací periody, vlhkého paliva a sníženého výkonu na emisní faktory výše uvedených škodlivin. Bylo zjištěno, že všechny tři výše uvedené vlivy způsobily podstatné zvětšení emisního faktoru sledovaných škodlivin, oproti emisním faktorům zjištěným při normálních provozních režimech kotlů.

1 INTRODUCTION

Emission inventories are utilizing emission factors (EF) for computation of source participation to total emissions. Domestic combustion has been identified as significant source of environmental pollution [1]. Emission factors of polyaromatic hydrocarbons (PAH), polychlorinated benzenes (PCBz), polychlorinated phenols (PCPh), polychlorinated biphenyls (PCB) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) during steady state regime of biomass and coal fuels combustion in four different types of domestic heating appliances were already published [2,3]. However, emission factors during non-stationary phases of combustion, combustion facility malfunction or during combustion of inappropriate fuels will be substantially higher. Effect of waste addition into the coal or biomass fuel were published previously e.g. by Hedman (2006) [4]. EFs of above-mentioned organic pollutants during selected other non-stationary states were determined and are presented in this paper.

2 MATERIALS AND METHODS

Two different fuels were used for combustion experiments: lignite and bitumenous coal. Results of ultimate and proximate analyses are depicted in Tab.1.

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Tab. 1 Results of ultimate and proximate analyses

	Lignite (L)	Bitumenous coal (BC)
Ash (wt. %)	4.18	3.01
Water (wt. %)	27.5	4.51
Combustibles (wt. %)	68.3	92.5
HHV (MJ/kg)	20.6	32.9
LHV (MJ/kg)	19.1	31.9
Carbon^d (wt. %)	64.7	81.7
Hydrogen^d (wt. %)	5.31	4.53
Nitrogen^d (wt. %)	0.897	1.06
Oxygen^d (wt. %)	22.6	11.0
Sulphur^d (wt. %)	0.855	0.702
Chlorine^d (wt. %)	0.00551	0.170

^{d} - in dry mass

Experiments were performed in three different types of boilers:

Boiler 1 - hot water boiler with over-fire concept of combustion, manual stoking and natural draft.

Boiler 2 - under-fire boiler with natural draft and manual stoking.

Boiler 3 - modern under-fire boiler with forced draft and automatic stoking by screw conveyor.

More details about tested boilers and resulted EFs obtained during steady state regime combustion runs were published elsewhere [3]. Summary of recently realized experiments and details about them are shown in Tab.2.

Tab. 2 Summary of realized runs

Boiler	Fuel	Run	Fuel feed rate (kg/h)	Output (kW)	Notes
1	lignite	1	8.1	21.6	3 h stoking period, stoke of 25 kg of lignite
		2–3	4.8	13.6	0.5 h stoking period, stoke of 2.5 kg of lignite
2	bitumenous coal	4	1.9	11.1	Wet fuel; water content in fuel ca 25 wt. %
		5–6	3.2	18.6	Dry fuel, water content in fuel 4.5 wt. %
3	lignite	7–9	5.9	23.9	100 % of nominal output
		10–12	3.7	15.4	65 % of nominal output
		13–15	1.9	8.0	33 % of nominal output

Boilers were tested at domestic combustion testing facility consisting of balance, tested boiler, isolated chimney system (height 4 m) exhausting to a dilution tunnel hood, dilution tunnel and fan. More details about testing facility and all advantages of dilution tunnel application were presented elsewhere before [5]. All sampling and analyses of flue gases were performed in the dilution tunnel. Content of total organic carbon (TOC) was analyzed by Multi-FID 100 by means of FID principle

(EN 12619). Particulate matter (PM) content was determined in accordance with ISO 9096. CO was measured by multi-gas analyzer Advance Optima by means of infrared principle and in accordance with EN 15058. Flue gas sampling and above-mentioned organic compounds analyses were performed in accordance with EN 1948. PAH is a sum of 10 compounds - fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene, dibenzo(a,h)anthracene, indeno(1,2,3-cd)pyrene. PCB is a sum of following congeners PCB No. 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 170, 180, and 189. TEQ PCB was calculated based on WHO-TEFs 2005. PCDD/F is a sum of Tetra- to OctaCDD/F, TEQ PCDD/F was calculated based on TEFs according to EN 1948.

3 RESULTS AND DISCUSSION

Effect of three different non-stationary states of combustion on emission factors of selected pollutants was observed: inappropriate fuel stoking on boiler with periodical operation, wet fuel due to inappropriate storage and automatic boiler operating on lower output than nominal. Above-mentioned states were chosen based on relevancy that these states could possibly occur during domestic heating. For comparison, combustion tests during steady state regimes were also realized.

3.1 Effect of stoking period

Boiler 1 with over-fire combustion concept is periodical boiler operating in dependence of length of stoking period. Therefore, the effect of stoking period and fuel batch size was observed on this boiler. Combusted fuel was lignite. Run 1 was realized with 3-hour long period and amount of stoked fuel was ca 25 kg of lignite (3/4 of storage). Runs 2–3 were operated with 0.5-hour long period, batches of fuel were ca 2.5 kg of lignite, length of whole test was 4 hours. Average fuel feeding rate decreased from 8.1 kg/h (output 21.6 kW) for run 1 to 4.8 kg/h (output 13.6 kW) for runs 2 and 3. Shortening of stoking period to 0.5 hour lead to 2–4 times decrease of EFs of particulate matter, CO, TOC and PAH, i.e. main products of incomplete combustion with no relation to chlorine. Emission factors of PCB and TEQ PCB increased ca. by 10–20 %. EF of TEQ PCDD/F was nearly the same. PCDF/PCDD ratio was different for runs 1 and 2–3. For run 1 PCDD were prevailing products (ratio 0.7), for runs 2–3 the PCDF/PCDD ratio was 1.5–2.0. The reason for this phenomenon consists probably in significantly higher levels of PCDD/F formation precursors (e.g. PCPh) during this run because formation via condensation of precursors is given for PCDD mainly. Moreover, homologue profile of PCDD/F during run 1 was shifted towards low-chlorinated PCDD/Fs due to lack of chlorination agent and increase of products of incomplete combustion. Obtained EFs of selected pollutants are shown in Tab.3 and Tab.4.

Tab. 3 EFs of particulate matter, CO, TOC and PAH, boiler 1

Run	PM (mg/kg)	CO (mg/kg)	TOC (mg/kg)	PAH (mg/kg)
1	37 800	150 000	57 000	160
2–3	10 100±500	73 900±3 600	16 200±500	38.9±15.2

Tab. 4 EFs of PCB and PCDD/F, boiler 1

Run	PCB (ng/kg)	TEQ PCB (pg/kg)	PCDD/F (ng/kg)	TEQ PCDD/F (ng/kg)
1	53.0	43.1	402	1.15
2–3	68.4±4.4	50.6±10.5	120±5	1.16±0.40

3.2 Effect of wet fuel

Effect of wet fuel was observed on boiler 2 with under-fire concept. These types of boilers consist of three parts - fuel storage, combustion chamber and gas flow chamber. Devolatilization and partial combustion occurs in a small part of fuel in the bottom of fuel storage, main combustion takes place in subsequent combustion chamber. Due to temperature gradient fuel drying process in fuel storage could be expected in this type of boilers so the water content in fuel does not need to have significant effect on EFs. Significantly higher EFs of non-chlorinated products of incomplete combustion like CO, TOC and PAH (2–3 times higher) can be concluded from Tab.5 for wet fuel (run 4). For PCB, PCDD/F, TEQ PCDD/F only small increase of EFs in comparison of run 4 and runs 5–6 was found (see Tab.6). No shift was found in homologue and congener profiles of PCBz, PCPh and PCDD. For PCDF increased content of low-chlorinated homologues was observed, it is partially in accordance with Shao [6] et al., however, they published increase also of low-chlorinated PCDD.

Tab. 5 EFs of particulate matter, CO, TOC and PAH, boiler 2

Run	PM (mg/kg)	CO (mg/kg)	TOC (mg/kg)	PAH (mg/kg)
4	16 700	189 000	62 900	140
5–6	7 810±950	107 000±7 000	23 200±700	62.7±15.5

Tab. 6 EFs of PCB and PCDD/F, boiler 2

Run	PCBz (μg/kg)	PCPh (μg/kg)	PCB (ng/kg)	TEQ PCB (pg/kg)	PCDD/F (ng/kg)	TEQ PCDD/F (ng/kg)
4	56.6	17.4	162	146	934	12.4
5–6	35.2±3.6	18.3±0.5	130±18	322±99	710±260	10.7±3.8

PCBz - sum of TriCBz to HxCBz, PCPh - sum of TriCPh to PeCPh

3.3 Effect of lower output

On modern automatic boiler with forced draft and automatic stoking by screw conveyor the effect of lower output than recommended nominal output was tested. Runs 7–9 were realized with nominal output 23.9 kW, fuel feeding rate was 5.9 kg of lignite per hour (fan output 90 %, screw conveyor on/off 12/20 s). Runs 10–12 were realized on ca. 65 % of nominal output, e.g. 15.4 kW, fuel feeding rate was 3.7 kg/h (fan output 45 %, screw conveyor on/off 6/23 s). Runs 13–15 were performed on 8.0 kW, e.g. 33 % of nominal boiler output, fuel feeding rate was 1.9 kg/h (fan output 7 %, screw conveyor on/off 5/40 s). High increases of EFs were obtained with boiler output decrease for all pollutants except PM. No increase of PM emission factors could be accounted to lower volumetric flow of air through boiler and thus the lower gas velocity. Deterioration in quality of combustion is the most evident from comparison of EFs from runs 7-9 and 13-15. EF of TEQ PCDD/F arises 10 times in case that boiler is operating on 33 % of nominal output. Homologue profile of PCDD/F is slightly shifted for runs 10-12 and 13-15 towards to higher-chlorinated PCDD/Fs, for PCPh and PCBz were these changes not observed. PCDF/PCDD ratio was similar for all runs which corresponds to the fact that content of the main condensation precursors of PCDD formation PCPh did not change significantly too. Generally, with decreasing output below the nominal “quality” of combustion also decreases and EFs of selected pollutants increase. The resulted EFs are summarized in Tab.7 and Tab.8.

Tab. 7 EFs of particulate matter, CO, TOC and PAH, boiler 3

Run	PM (mg/kg)	CO (mg/kg)	TOC (mg/kg)	PAH (mg/kg)
7–9	889±86	8 580±1 700	135±8	0.210±0.140
10–12	476±13	10 800±200	225±22	0.413±0.137
13–15	673±71	19 600±2 300	862±243	8.1±3.2

Tab. 8 EFs of PCB and PCDD/F, boiler 3

Run	PCBz (ng/kg)	PCPh (μg/kg)	PCB (ng/kg)	TEQ PCB (pg/kg)	PCDD/F (ng/kg)	TEQ PCDD/F (ng/kg)
7–9	33.7±9.5	-[a]	8.62±0.75	4.46±0.72	6.59±1.93	0.0740±0.0306
10–12	11 000±2 600	1.49±0.57	79.4±23.9	18.8±4.7	23.9±5.3	0.248±0.106
13–15	22 000±700	2.70±1.19	69.5±16.3	22.5±8.0	41.1±13.3	0.731±0.281

[a]- not determined due to matrix effect, PCBz - sum of TeCBz to HxCBz, PCPh - sum of TriCPh to PeCPh

4 CONCLUSIONS

It can be concluded that all observed non-stationary states (inappropriate stoking of fuel, wet fuel and boiler operating on lower output than nominal) have deteriorative effects on quality of combustion. Therefore, EFs of almost all selected pollutants increased during these states. Moreover, it was observed that only operating of boiler on lower output than nominal caused significant increase of EFs of chlorinated products of incomplete combustion like PCDD/F. Effect of boiler output was observed on modern boiler with semi-continuous automatic stoking, for boilers with manual batch stoking this negative effect would be even more significant. Therefore, connection of boiler with accumulation tank will results in substantial decrease of pollutants emissions because boiler can be operated with nominal output.

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