XYLOCLEAN: Combustion regulation and treatment of smoke from a wood stove

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Abstract:

In order to reduce pollutant emissions, a commercial wood log stove with staged combustion was fitted with an exhaust gas after treatment device based on a metal-based oxidation catalyst coated with platinum and palladium followed by a sintered metal particle filter (both used on diesel internal combustion engines) and a regulated exhaust gas fan for draft control. The tests were performed with standardised logs (wood species, size and humidity) precisely positioned in the furnace. Further, an exhaust gas draft control based on the pressure at the stove outlet enabled achievement of better operating stability than draft control based on the temperature. Regarding the exhaust gas emissions, the use of a catalyst lead to a complete (i.e. 100%) carbon monoxide (CO) oxidation, independently of both its internal structure and length, as well as the draft control parameter (pressure or temperature). In the case of draft control by pressure, the catalyst resulted in a significant reduction of polycyclic aromatic hydrocarbons (PAH) by 65 to 90 %, increasing with the cell density. The addition of a particle filter downstream had the same influence on particles emission as an increase in cell density. On the other hand, the exhaust gas temperature at the level of the catalyst was too low to permit significant methane (CH_4) oxidation and reduction of nitrogen oxides (NO_x). The use of a particle filter enabled a minimum of 90 % reduction of the mass emitted. However, this also lead to an important increase of the pressure losses between the stove and the fan inlet, resulting in higher electricity consumption. Globally, an active combustion control by draft regulation combined to an exhaust gas after treatment based on oxidation catalyst and particle filter appears to be a promising solution to reduce wood log stove exhaust gas emissions and should be further studied.

Keywords:

Wood log stove, Wood combustion, Exhaust gas after treatment, Particle filtration, Draft control, PAH emissions.

1. Introduction

In a general way, the promotion of renewable energy use leads to a constant increase in the number of wood fuel heating systems. Due to the difficulty to limit the exhaust gas emissions of small wood stoves (< 70 kW), the Swiss authorities, like numerous other countries, prescribe less stringent limit values than for large installations (> 70 kW) [1].

The emissions of fine particles from these wood combustion appliances are several times higher than those from gaseous and liquid fuel ones, which however, taken all together, produce considerably more heat and are similar to transport in their total emissions [2]. As has already been scientifically acknowledged in numerous publications, these particles can directly damage health, depending on their size and chemical properties.

The majority of wood heating systems are fed manually. Their emissions depend not only on the combustion technology involved but also on the conditions imposed by the user. The wood moisture, the ignition conditions, the way of loading the system and the draft setting can affect the particulate emissions by a factor higher than 100 [3]&[4]. Further, if the installation is connected to a natural draft chimney, the particulate emissions can vary considerably during log combustion due to atmospheric changes. This detrimental operating variability, when occurring in winter while

numerous small wood heating systems are on duty, contributes significantly to the frequent exceedance of the particulate matter immission limit (PM10).

Consequently and despite the absence of particulate emission limit for small devices, it appears necessary to reduce the environmental impact of small manually loaded wood heating installations. Exhaust gas after treatment systems are already available on the market [5][6][7][8]. However, they are characterised by a limited efficiency and/or generate critical residues (ash or sludge), which have to be eliminated. In order to achieve higher efficiency, lower cost and to palliate the disadvantages of actual systems, further development of exhaust gas after treatment systems is required. Similar systems have already been successfully implemented on internal combustion engines, where they have achieved high efficiency at very low cost due to mass production. The availability of automotive components, like catalytic converters or particle filters, motivated the authors to study their potential to reduce exhaust emissions from wood combustion. This motivation was also partly supported by a former study on the use of an oxidation catalyst [9], which indicates that a platinum/palladium coating, widely used in engine application, is particularly efficient for reducing CO and VOC emissions.

The main goal of this study was to evaluate different exhaust gas after treatment systems, based on catalytic converters (CC) followed by a particle filter, on a commercial wood log stove with staged combustion. This art of combustion is mainly used in wood boilers, in order to achieve very low raw emission. The choice of the wood stove type and the CC position, in front of the filter, has been made in order to limit the risk of filter plugging by condensation of heavy oxygenated hydrocarbons generated during poor wood combustion.

2. Methods

2.1. Equipment used

2.1.1 Test bench arrangement

Fig. 1 shows the stove used equipped with a catalytic converter and a particle filter. The exhaust flow rate is measured with a Venturi tube and a controlled fan regulates the draft
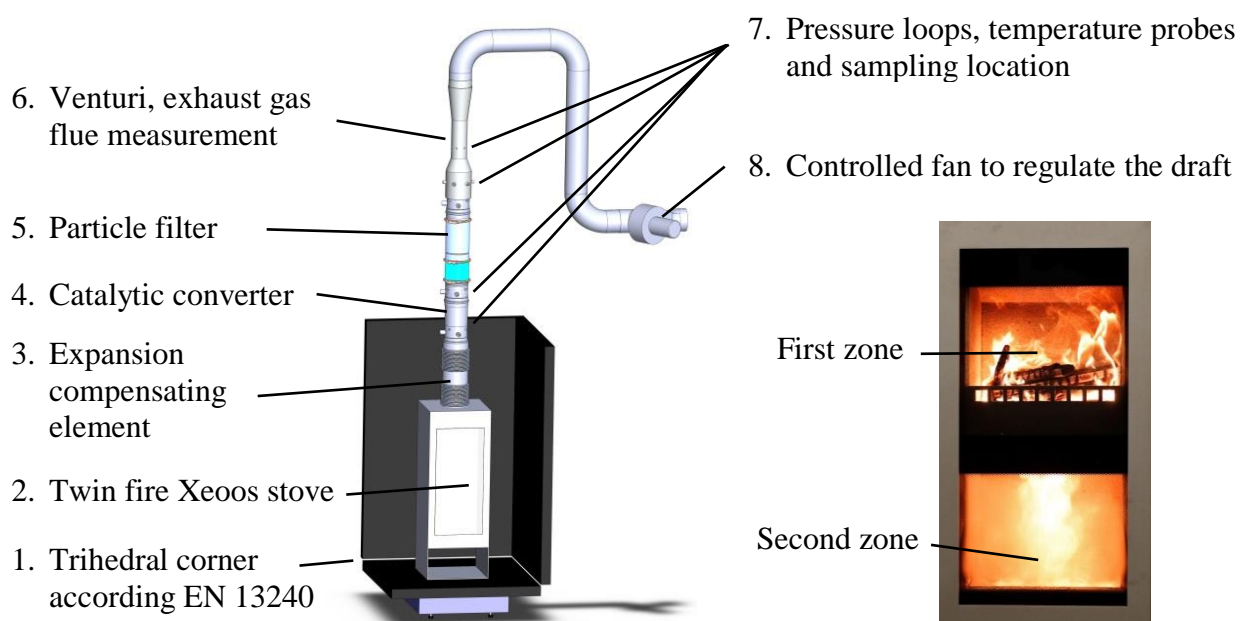

7. Pressure loops, temperature probes and sampling location

6. Venturi, exhaust gas flue measurement

8. Controlled fan to regulate the draft

5. Particle filter

4. Catalytic converter

3. Expansion compensating element

2. Twin fire Xeoos stove

1. Trihedral corner according EN 13240

Fig. 1 - Test bench arrangement.

First zone

Second zone

Fig. 2 - Staged combustion wood stove in steady state operation.

The components related to **Fig. 1** are described in the following sections.

2.1.2 Stove Xeoos (2)

The stove used is manufactured by the company Specht GmbH & Co. KG, Hatzfeld, Germany. Its nominal power rating is 5 kW. It is designed with two combustion zones. The log is loaded in the first zone, on the upper half of the stove. There is a handle to toggle from the starting mode (first zone only) to the steady state mode (both zones used, with post combustion in the second zone). The tests were only carried out in steady state mode (**Fig. 2**). No modification were performed on the stove.

2.1.3 Draft control (7)

A fan (ref. RG1 R-2D) manufactured by FITZE VENTILATOREN AG (Untervaz, CH), with frequency converter PID, controls the draft on basis of suction pressure or stove outlet temperature.

2.1.4 Catalytic converter (4)

Four different catalytic converter substrates (two types of structure and two lengths) were provided by Emitec GmbH at Lohmar (D) and coated with platinum/palladium by Umicore AG + CO.KG, Hanau-Wolfgang (D); see **Table 1** and **Fig. 3, Fig. 4 & Fig. 16.**

Table 1- Catalytic converter specifications.

#	Coating [qty/type/ratio] *	cell [cpsi] / thickness [mm] - type **	Dim. ($\varnothing \times$ h) [mm]
C1	150 g.ft^{-3} Pt/Pd 1/1	300 / 0.04 - standard	150×74.5
C2	150 g.ft^{-3} Pt/Pd 1/1	300 + 600 / 0.04 - LS	150×74.5
C3	150 g.ft^{-3} Pt/Pd 1/1	300 / 0.04 - standard	150×120
C4	150 g.ft^{-3} Pt/Pd 1/1	300 + 600 / 0.04 - LS	150×120

* g.ft^{-3} = gram per cubic foot
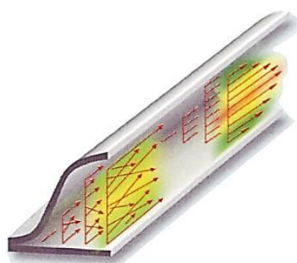** cpsi = cells per square inch, thickness of the support

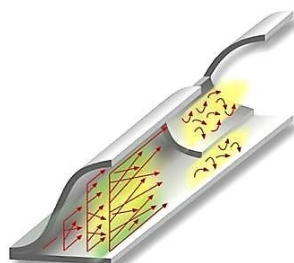*Fig. 3 - Catalytic converter, standard structure.*

Fig. 4 - Catalytic converter, LS structure.

Fig. 5 - Particle filter.

The detailed characterisation of the catalyst was subcontracted to the Paul Scherrer Institute[1] (PSI). The main results are:

- The conversion of the CO is greater than 99.5%, above 150°C;
- The complete conversion of the CH_4 needs a temperature around 450°C. There is already a significant conversion (between 20 and 30%) above 350°C;
- The highest molecular weight hydrocarbons, like C_3H_6 (propene), are converted (> 99.5%) above 150°C into other kinds of hydrocarbons, which might deposit on the catalyst front face. From 250°C, they are directly converted to CO_2 (99%);
- The total conversion of the naphthalene occurs only above 150°C. It is necessary to reach a temperature of 250°C to avoid deposit of secondary hydrocarbons on the catalyst;
- Test on a mixture of gas (CO, NO and C_3H_6) showed a total conversion above 200°C and a fast reduction of the ratio of conversion below 200°C;
- The length of the structure of the catalytic converter does not have a significant influence. This is probably due to its high platinum/palladium content.

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[1] Paul Scherrer Institute, Switzerland, www.psi.ch

2.1.5 Particle filter (5)

HJS, Menden (D) provided the sintered metal particle filter. The filtration surface area is 2.7 m^2 and its diameter is 150 mm (**Fig. 5**). Its stated effectiveness is more than 99% for particles from 20 to 300 nm.

2.2. Instrumentation

2.2.1 Pressure

Pressure is measured by the means of four loops: pressure losses across the catalytic converter and the particle filter, as well as pressure difference at the Venturi. The sensors are made by Halstrup Walcher, with a range of ± 50 Pa and a precision of 0.3 Pa.

2.2.2 Temperature

Temperature is measured with thermocouples type K (class 2), at the outlet of the stove, after the catalytic converter and after the particle filter.

2.2.3 Control

The complete system is controlled with a Labview® based software. It is possible to select the command mode based on the pressure or on the stove outlet temperature. The data are recorded every second.

2.2.4 Particulate sampling system

The particulate were collected, according to EN 13284-1 standard, by an Automatic Isokinetic Sampling System (ITES) from Paul Gothe GmbH (D) fitted with Munktell filter media, grade MG160 (**Fig. 6**).

The quantity of particles is the differential mass of the filter before and after sampling. The filters were prepared according the FOEN[2] [10] and weighed using an analytical balance XP105DR from Mettler Toledo, with a range of 0...31 g / 120 g and a precision of 0.01 mg / 0.1 mg.

2.2.5 Exhaust gas condenser

The gas extracted during particles sampling is condensed at −25°C (**Fig. 6**). The condensate is analysed by chromatography to determine its polycyclic aromatic hydrocarbons (PAH) content (see 2.3.2).
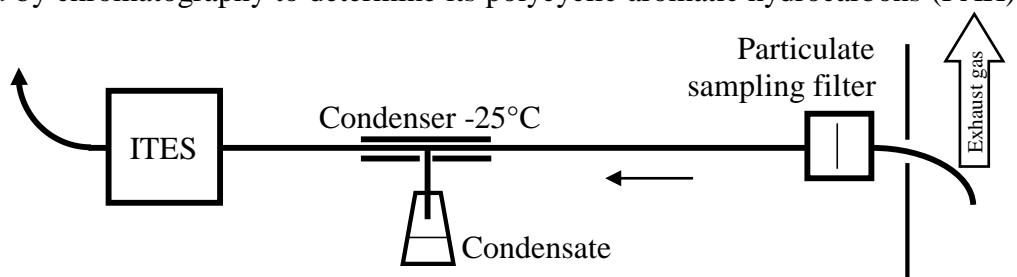

Fig. 6 - Particulate sampling and gas condenser installation

2.2.6 Exhaust gas composition

The exhaust gases were analysed every second with a gas analyser MRU/MGA5 as follows:

- Exhaust gas temperature (precision: 1% if > 200°C) and ambient air (± 1°C);
- Atmospheric pressure or differential pressure (1% of the value);
- O_2 (± 0.2 Vol. % abs.), CO_2 (0 to 15.85±0.48 Vol. % abs.), CO (0 to 821±25 ppm), NO (0 to 404±12 ppm), NO_2 (with an NO converter) and total hydrocarbons by CH_4 equivalent (0 to 794±24 ppm).

2.2.7 Wood log characteristics

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[2] FOEN: The Swiss **F**ederal **O**ffice of **EN**vironment is the centre of environmental expertise of the Federal Department of Environment, Transport, Energy and Communication.

The log characteristics were determined according to EN 13240 [11].

In order to limit the inter-experimental variation and improve the reproducibility, each load contained three calibrated logs to obtain an average combustion of one hour (**Table 2** & **Fig. 7**).

The total volume of wood needed for the whole experiment was about one cubic meter.

Table 2 – Calibrated log characteristics.

Subject	Data
Type of wood	Beech
Log dimensions	$60 \times 60 \times 200$ mm
Number of logs per load	3
Load weight	1356 ± 8 g
Moisture (wet base)	$15.2 \pm 0.3\%$
Gross heat (dry base)	19776 ± 160 kJ.kg^{-1}
Ash	$0.40 \pm 0.09\%$

Fig. 7 - Calibrated wood log arrangement on dedicated combustion holder.

2.3. Testing

2.3.1 Description

For each test, the positioning of the calibrated logs (**Fig. 7**) were the same. The first test started only after three loads with non-calibrated logs, to warm up the entire installation to the right temperature (210°C) and to build up an ember bed. Each standard test was made in steady state (**Fig. 2**).

All the wood used, calibrated or not, and the ash produced were weighed for each test to determine the final combustion residues.

Particle sampling with the ITES device started right at the beginning of a test and continued during one hour (duration of a complete combustion).

Filters used to collect particles were dried for four hours at 160°C and stored before weighing at ambient temperature in a desiccator.

Two modes were available to pilot the combustion: the first one by temperature and the second one by pressure. The temperature probe (resp. pressure) used for the control was placed just above the expansion compensating element (**Fig. 1; 3**).

2.3.2 PAH analysis method

The Institute of Chemical Technology of HEIA-FR analysed the PAH content of the exhaust gas. The PAH are molecules composed of at least two cycles of benzene (**Fig. 8**). The benzene is the main component of PAH and is composed of six carbon atoms.
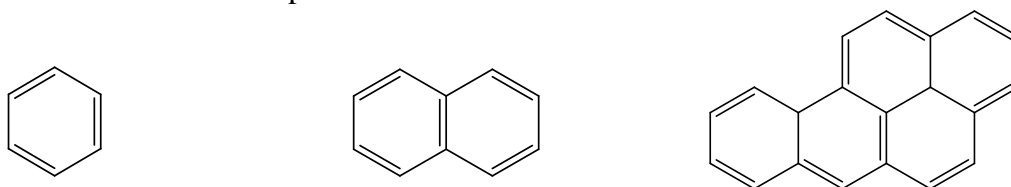

Fig. 8 - Example of PAH, from left to right: benzene, naphthalene, benzo[a]pyrene.

They are produced if there is an incomplete combustion of coal, oil, wood, etc. and they are found in the ambient air making them a good tracer of the quality of the combustion.

The sixteen PAH analysed, ranked by toxicity in the 1980s by the US Environmental Protection Agency are considered as priority pollutants (**Table 3**).

Table 3 – List of the 16 priority PAH analyzed by HEIA-FR.

PAH	Formulation	CAS #	Toxicity

Benzo(a)pyrene	$C_{20}H_{12}$	50-32-8	Human carcinogen
Dibenz(a,h)anthracene	$C_{22}H_{14}$	50-70-3	Probable human carcinogen
Benz(a)anthracene	$C_{18}H_{12}$	56-55-3	Probable human carcinogen
Benzo(b)fluoranthene	$C_{20}H_{12}$	205-99-2	Probable human carcinogen
Benzo(k)fluoranthene	$C_{20}H_{12}$	207-08-9	Probable human carcinogen
Chrysene	$C_{18}H_{12}$	218-01-9	Probable human carcinogen
Indeno(1,2,3,c,d)pyrene	$C_{22}H_{12}$	193-39-5	Probable human carcinogen
Benzo(g,h,i)perylene	$C_{22}H_{12}$	191-24-2	-
Anthracene	$C_{14}H_{10}$	120-12-7	-
Acenaphtene	$C_{12}H_{10}$	83-32-9	-
Acena phtylene	$C_{12}H_{18}$	208-96-8	-
Fluoranthene	$C_{16}H_{10}$	206-44-0	-
Fluorene	$C_{13}H_{10}$	86-73-7	-
Naphtalene	$C_{10}H_8$	91-20-3	-
Phenanthrene	$C_{14}H_{10}$	85-01-8	-
Pyrene	$C_{16}H_{10}$	129-00-0	-

2.3.2.1 Analysis method

Method to obtain the PAH sample is a liquid-liquid extraction followed by gas chromatography (GC) coupled with mass spectrometry (MS) based on the method developed by the FOEN [12][9].

2.3.2.2 Equipment

- Device: GC Trace with MS ISQ, from Thermo SCIENTIFIC;
- Column: DB5-MS / 30m x 0.25mm x 0.25um;
- Vector gas: Helium with constant flow at 1.5 ml/min;
- Settings of the oven: 40°C for 4 min, 30°C/min until 100°C, 10°C/min until 260°C, 2°C/min until 330°C and finally 3 min à 330°C;
- Injector: 280°C, injection 1μl in splitless mode;
- MS: Sample transfer line at 280°C, source at 200°C, SIM at m/z = 128, 152, 153, 166, 178, 202, 206, 228, 252, 254, 276, 278.

2.3.2.3 Products used

- PAH std.: Standard Mix of the 16 PAH / Supelco Analytical Batch # LB59411;
- Internal std.: 2.2'-binaphtyl / Sigma-Aldrich Batch # S413402;
- Recovery std.: indeno(1,2,3-cd)fluoranthene / Certified Reference Material Batch # BCR-267.

2.3.2.4 Sample preparation

Exhaust gas is condensed as mentioned in 2.2.5 at -25°C, to capture PAH. After each test is completed, all of the instruments are cleaned with three volumes of dichloromethane: Condensate and solvent are stored in 100ml volumetric flasks.

After adding the internal standard (see above), the sample is transferred into a separating funnel of 100 ml and extracted two times 20ml of dichloromethane to separate aqueous and organic phase from one another. Organic phase is concentrated at 5ml in a rotary evaporator.

The sample is transferred into a 10 ml volumetric flask, the recovery standard is added and the total is filled to the mark with dichloromethane.

2.3.2.5 GC analyses

One microliter of each standard Mix of the 16 priority PAH (C_{total} = 0.5 ppm / 2 ppm / 5 ppm / 10 ppm) are injected in the GC-MS to determine their relative retention factors and a curve of calibration is drawn for one of each.

After that, one microliter of each sample is injected in the GC-MS and by comparison with the calibration curves, the total sum of the 16 PAH is calculated.

2.3.2.6 Results conditioning

For each sample, the total amount of the 16 PAH is given in ppm (mg.l^{-1}).

The quantification limit of the method for the total sum is 0.15 ppm**.**

2.3.3 Experimental plan

Four catalytic converters, numbered C1 to C4 (table 1) and one particle filter were used. Nine configurations (A to I, **table 4**) were designed and each one was repeated three times.

Configurations A and B are the references, without treatment. It means that there is no catalytic converter nor particle filter. Only the control method was varied: by pressure (-12 Pa, according EN 13240) for A and by temperature (210°C, standard temperature according manufacturer) for B.

Preliminary tests showed that the control by pressure induced a higher temperature (240°C) than operating temperature stated by the manufacturer. However, the nominal operating value of the manufacturer (210°C) was maintained for the temperature control mode.

Control by temperature does not result in a better efficiency and seems to produce more particles. Nevertheless, it was decided to use it in configurations C and I for comparison with D and H.

In order to reduce the number of test, the particle filter was used only for two configurations (control by pressure and temperature) after the CC. The configurations H and I were defined when results from configurations C, D, E, F and G were known. The C2 was selected because it has the best results for the particle emission without the particle filter and has for the other pollutants similar results than the longest ones (C3 and C4) and better results than the C1.

The CC is positioned just above the stove to increase its efficiency (with the temperature of the exhaust gas) and to minimize soot and tar on the particle filter.

Table 4 - configurations tested

Configuration	Catalytic converter (see table 1)	Particle filter	Control by:
A	No	No	pressure: -12 Pa
B	No	No	temperature: 210 °C
C	C1	No	temperature: 210 °C
D	C1	No	pressure: -12 Pa
E	C2	No	pressure: -12 Pa
F	C3	No	pressure: -12 Pa
G	C4	No	pressure: -12 Pa
H	C2	Yes	pressure: -12 Pa
I	C2	Yes	temperature: 210 °C

3. Results and Discussion

Calculations were made according EN 13240 [11].

The results are considered as significant if the t-test statistic is positive with an $\alpha = 5\%$.

The confidence interval of 95% is represented in red on all the following graphs.

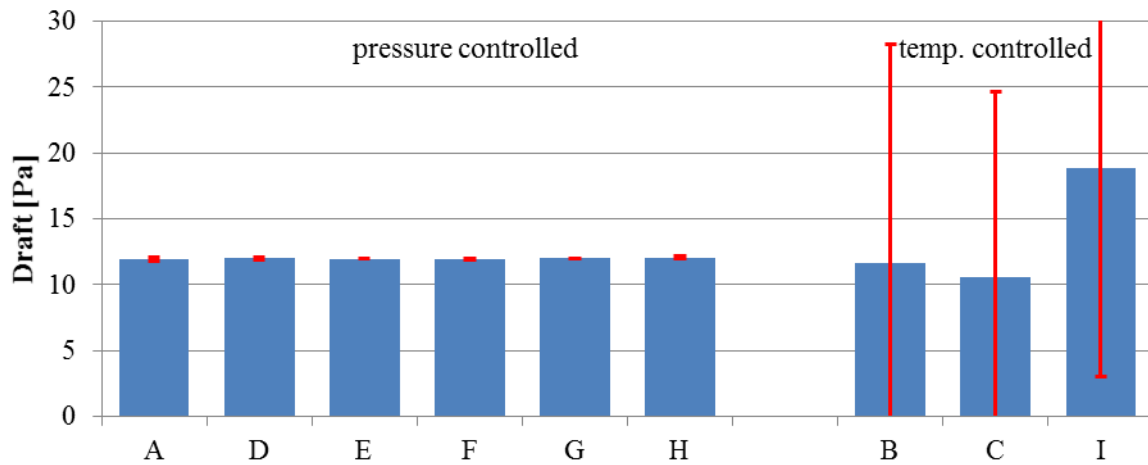The reference configuration for pressure and temperature control are the configuration A and respectively B.

Fig. 9 - Draft with confidence interval.

In **Fig. 9**, there is a large variability in pressure values when the system is controlled by temperature. With this mode, the regulating fan (**Fig. 1; 7**) works to maintain the temperature set point. Even if the draft is lower for C than B, the difference is not significant. The control by pressure, at -12 Pa at the stove outlet, in accordance with the EN 13240 standard is easily repeatable.

In order to compare the results between experiments, the pressure must be maintained at -12 Pa. Depending of the configuration, an increase in the fan motor frequency from 15 Hz to 30 Hz is necessary to maintain -12 Pa. Unfortunately, this control mode does not represent real installation; most stoves used do not have a draft control.

On **Fig. 10**, the pressure loss was measured on separated element: catalytic converter and particle filter, with the difference of pressure before and after the device studied. The total pressure loss for configuration H (resp. I) is the sum of both (particle filter and CC). To know the partial pressure loss of the particle filter alone, these results have to be subtracted from E (resp. C).

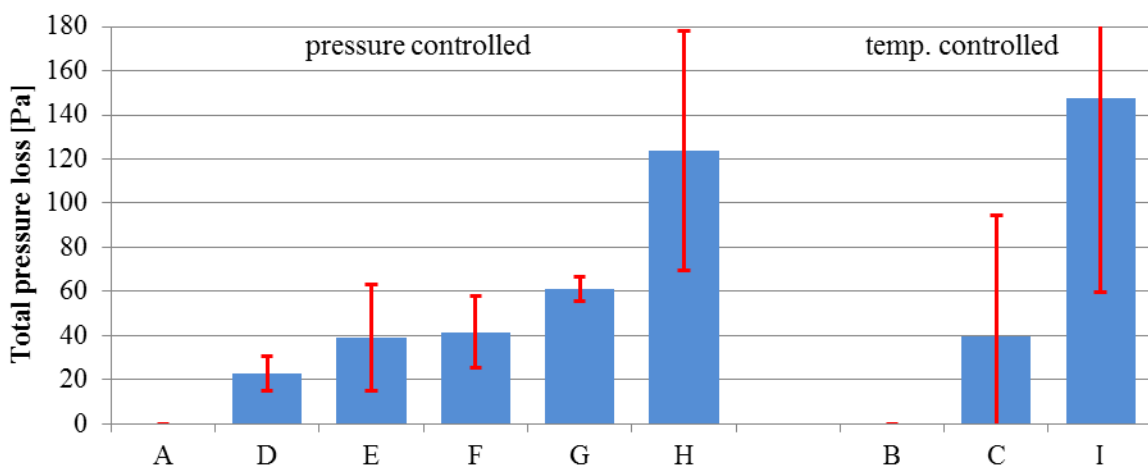Deposit on the particle filter surface generates a high variability: 124±54 Pa (H) and 148±88 Pa (I).

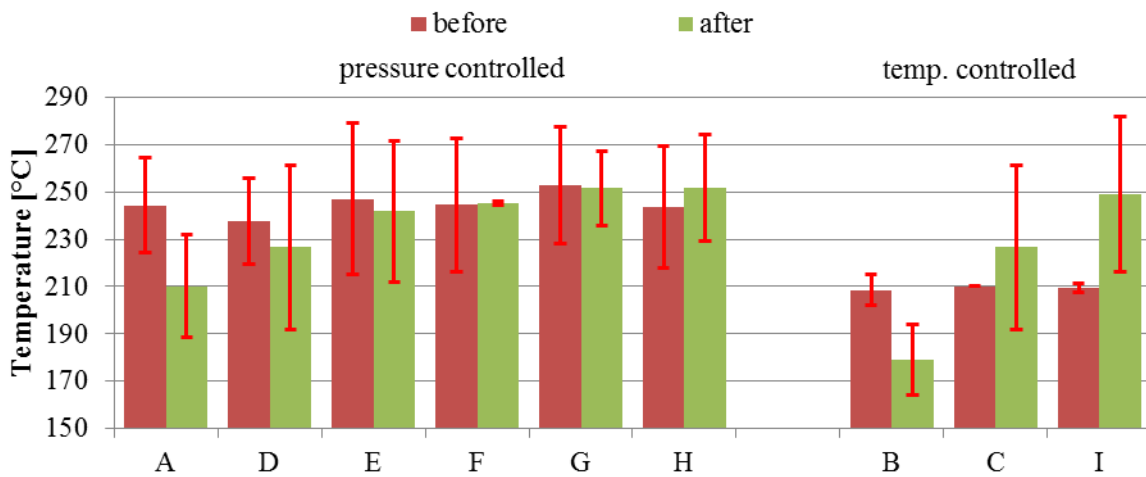*Fig. 10 - Total pressure loss generated by the after treatment device.*

Fig. 11 - Fume temperature before and after catalytic converter.

The temperature difference before and after the catalytic converter (**Fig. 11)** is variable according to the type and the efficiency of the CC but for all configurations, the difference of temperature after the CC is significant compared with A and B. With the configurations A and B, with no treatment, the temperature naturally decreases; as there is no insulation (an empty pipe replaces the CC). Its dispersion before the CC is higher when the control is made by pressure than temperature but similar after. The use of a CC reduces the gap. The oxidation is an exothermal reaction and when there is an important production of CO and PAH (config. C & I), the temperature increases. It is the case of the configurations H & I too but with a higher value due to the particle filter pressure loss.

The catalytic converter oxidizes totally the CO, regardless of the type used (**Fig. 12**). For the hydrocarbons equivalent (CH_4) and the NOx, there is no significant changes (**Fig. 13**). The PSI study showed that it is not possible to oxidize CH_4 under 350°C (totally at 450°C). It is not possible to reach this kind of temperature in the chimney so it would be necessary to study the possibility to place the CC in the stove near the outlet.

For the PAH emissions (**Fig. 14**), the variability for the configuration B (temperature control) is high: 8.4±13.4 ppm. All catalytic converters are efficient (between 67 to 96% with pressure control and between 85 to 98% with temperature control). For the configurations F to I, values are below the limit of quantification (0.15 ppm). It was noticed that the particle filter has a passive action on the quantity of PAH by collecting the rest of soot and tar that contain mainly theses pollutants (comparison between the configurations C and I).
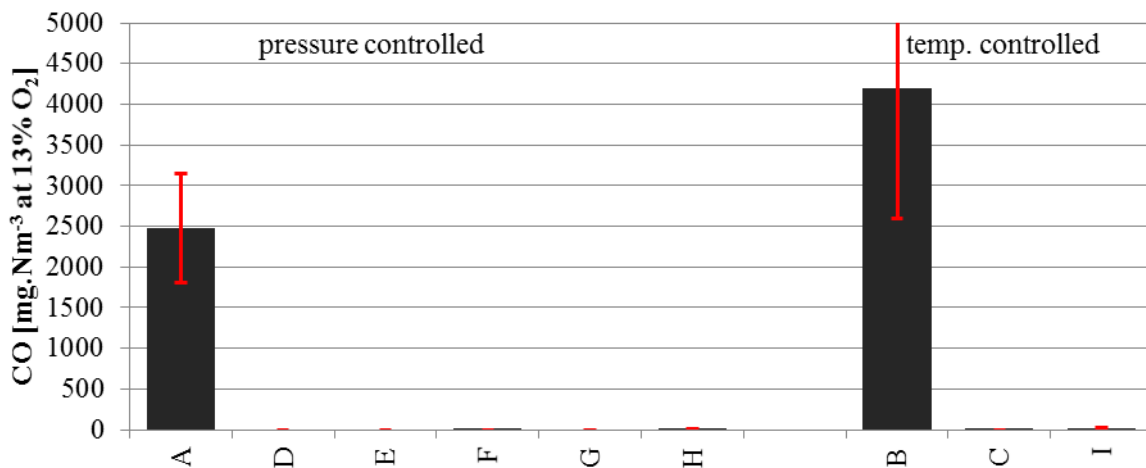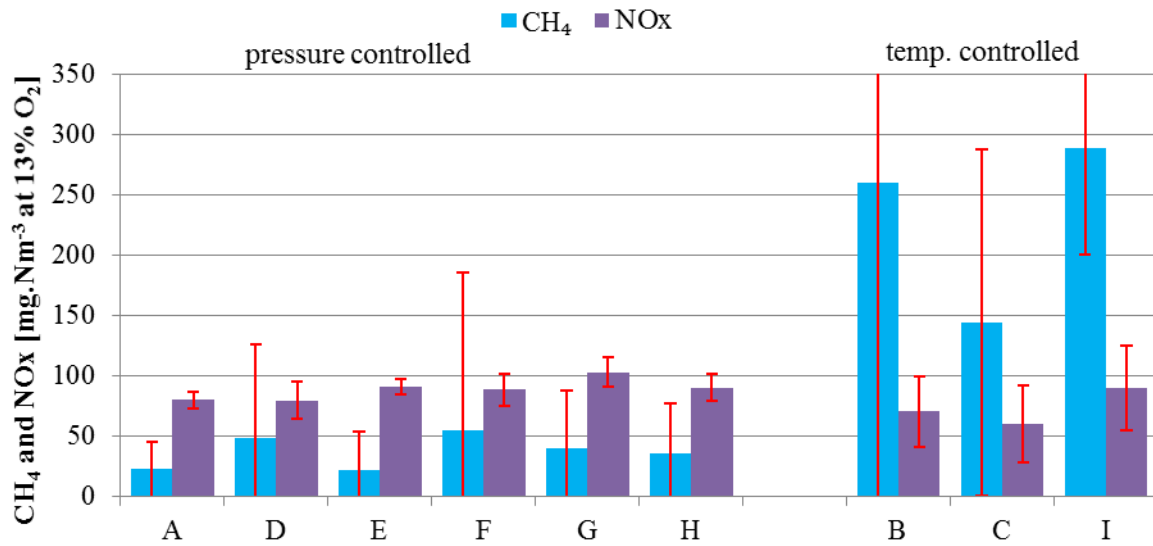

Fig. 12 - CO emissions.

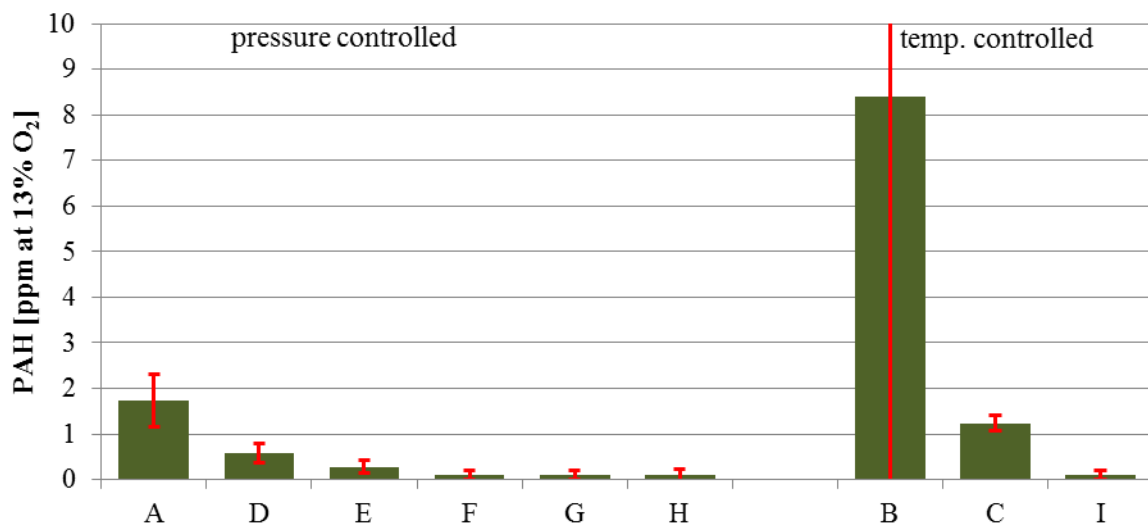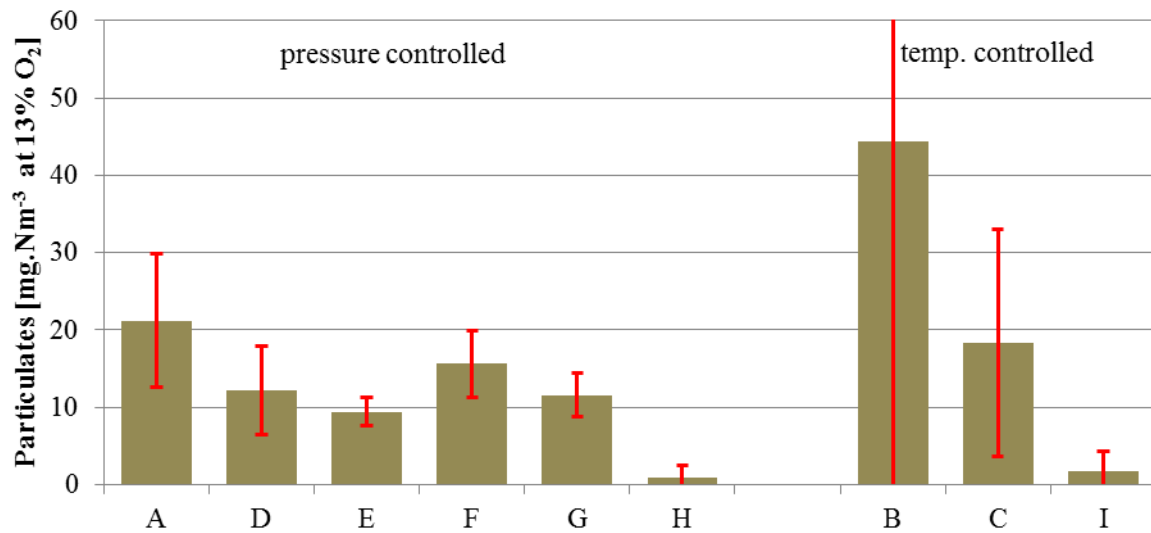*Fig. 13 - Pollutant emissions – CH₄ and NOx.*

Fig. 14 - PAH emissions.

Fig. 15 - Particulate emissions.

For the particulate emissions, the variability with the B configuration is also very high: 44 ± 56 mg.Nm^{-3} (**Fig. 15**).

The use of a catalytic converter significantly reduces particulate emissions and variability.

The catalyst operates as a "filter" and a deposit is progressively building up on the inlet surface (**Fig. 16**). After an average of 19 combustions, it is necessary to clean the surface with a stream of air at low pressure. A complementary study with the PSI indicates that 22% of the mass of this deposit is black carbon.
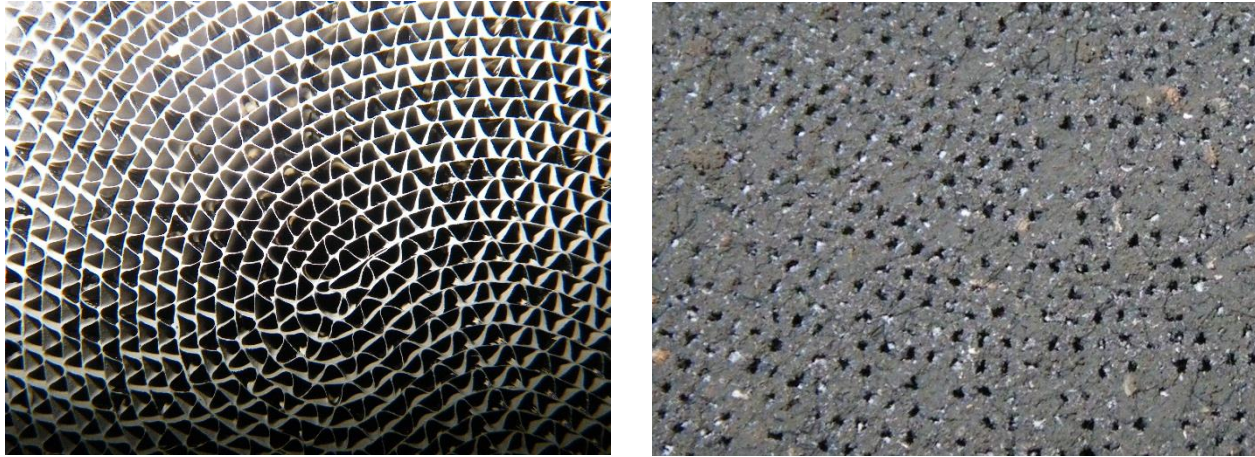

Fig. 16 - Deposit in front of catalytic converter C2 compared with the clean surface (left)

The particle filter significantly reduces particles, although with a high-pressure loss especially with the temperature control mode. Electricity consumption increases with increasing pressure loss. Electricity consumption was not recorded in this study, just measured punctually.

Using only a catalytic converter, the weight of particles emitted is 9 ± 2 mg.Nm^{-3}. This is less than half of the value stated by the manufacturer.

The configurations E and H (resp. C and I) are composed with the same catalyst but E is without the particle filter (**Fig. 15**). The addition of the particle filter (in H and I) decreases drastically the particulate emissions.

Using a particle filter is effective but it should be studied in another configuration (e.g. location, type of filter…) to limit the pressure loss.

We noticed that the choice of the mode of control (pressure or temperature) leads to large differences on pollutant emissions. The pressure control is better but the temperature control mode is interesting at the beginning of the combustion to quickly obtain a steady state and reach the optimal temperature for the catalytic converter.

An optimized installation is expected to combine both modes.

4. Conclusions

The main results of this study are:

▪ The stability of the combustion is better with the pressure control mode of -12 Pa at the outlet of the stove than with the temperature control mode at 210°C;

▪ The catalytic converter can completely (100%) convert the CO to CO_2 regardless of its structure or length, as well as of the control mode used.

▪ In the case of draft control by pressure:

▫ The catalytic converter converts PAH significantly (65 to 95%) especially if the cell density of is high. The addition of a particle filter has the same positive effect as the densification of the

cells. However, the temperature reached in the catalyst is not high enough to significantly reduce methane (CH_4) emissions;

- The catalytic converter with the LS structure an apparent density of 600 cells per square inch (cpsi) and a length of 74.5 mm led to the best compromise between the reduction of pollutant, the pressure loss and the investment based on the coating with precious metal;
- With a particle filter, it is possible to reduce more than 90% of weight of particles. However, the use of a particle filter increases pressure loss and therefore electricity consumption.

Therefore, a draft based active control of the combustion combined to a passive exhaust gas post-treatment (catalytic converter + particle filter) appears to offer a promising solution to reduce pollutant emissions from wood stoves.

5. Acknowledgment

6. References

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