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Physicochemical characterization of particles emitted during the three phases of wood logs combustion in domestic appliances

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Abstract

The fossil resources depletion led the world to use increasingly renewable energy. The first renewable energy is the wood energy, especially the domestic one. This energy transition has to be realized while taking account every environmental issues, like the air quality. Nevertheless, the domestic wood energy is one of the most important sources of fine particles emissions. In this study the particles emitted by each phase of the combustion of a log were characterised. The Total Suspended Particles, the granulometric and the organic and elemental carbon composition of the particles were followed.

Keywords: Biomass; Energy; Log Stove; Particles; OC/EC; SEM; EDS

1. Context

Ambient particles have a significant part in respiratory diseases (Löndahl et al., 2008). They have a significant impact in the modification of the radiative balance of the earth (IPCC, 2007) and the chemical composition of the clouds (Piot, 2011). In order to reduce risks, the most important sources of emissions have to be targeted and residential wood energy was assessed in many studies as a main source. Despite the fact that the emission factor depends not only on the kind of sampling but also on the country, wood energy is an important source of particles, particularly in log stoves (Pettersson et al., 2011). Many studies defined the toxicology and the cytotoxicology of the particles emitted by wood

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combustion (e.g., Nussbaumer et al., 2005; Tapanainen et al., 2011; Torvela et al., 2014). In a log stove, the combustion can be divided in three parts: ignition, homogeneous combustion and heterogeneous combustion (Loo Van, 2002). In order to better understand the effects of the particles on the health, the characterization of the different particles emitted must be achieved. The first step of this challenge aim to know if the particles emissions are constant during the combustion and if they are not, to define which kinds of particles are emitted during each phase of combustion.

2. Materials and methods

A log stove of 6 kW (2012) was used for the experimentations. This log stove possessed a primary and a secondary air supply and an isolated combustion chamber. The applied fuel was beech logs with a moisture content of 12% (EN 13240). The logs were selected without nodes and bark to get a higher repeatability. The Wet Low Calorific Value (LCVw) was 16700 kJ kg^{-1}. All the experimentation was realized with the same load (about 1.8 kg) composed by two logs of similar masses and was repeated three times.

The O_2 and the CO emissions were continuously measured by photochemical cells. In order to avoid an over/under estimation of the global emissions, the temperature for every sampling of particles or Volatile Organic Compound (VOC) was hold at 160 °C. As the aim of this study was to associate the particles emitted with the different steps of the combustion, no dilution was applied in order to maintain the chemical composition of the particles (Lipsky et al., 2006). The VOC and the CH_4 emissions were characterized with a Flame Ionization Detector (FID). The Total Suspended Particles (TSP), Total Carbon (TC), Organic Carbon (OC) and Elemental Carbon (EC) were sampled on quartz filters with an isokinetic probe. The TC, OC and EC composition of the particles were performed by a thermal-optical analyzer (Sunset Laboratory Inc.). In order to characterize the granulometric repartition of the particles, an Electric Low Pressure Impactor (ELPI) was sampling continuously the smoke in the duct.

The elemental composition of the particles was determined using a Field Emission Gun Scanning Electron Microscope (FEG SEM) equipped with an Energy Dispersive Spectrometry (EDS). The sampling was performed during 25 seconds with a sampling flow of 1 NL min^{-1} on a polycarbonate filter with a porosity of 200 nm. The module “Feature” of the software “INCA” (Oxford-Instruments) allowed an automated detection and a micro-analysis of the particles. In order to get a consistent representativeness, 2000 particles were analyzed per filter. Even if the EDS is qualitative, this analysis procedure presented a good repeatability (Lettino & Fiore, 2013). Thanks to this micro-analysis, some groups of particles with a specific composition were defined.

3. Results and discussions

3.1 Results on the gaseous and particles emissions

As expected, the three phases of the combustion of a log do not emit the same quantity of pollutants. Table 1 presents all the results concerning the gaseous emissions. During the ignition, CO, VOC and CH_4 are high concentrated in the smoke. This is mainly due to a high volatilization of pyrolysis gases which led to a low O_2 rate in the smoke. Then, the meeting statistics between the pyrolysis gases and the O_2 are low. The homogeneous combustion is the less emissive step and the heterogeneous combustion is the more emissive step. These high emissions are caused by a decrease of the temperature and the

disappearance of the flame. Moreover, a log stove is designed to burn gases and is less performant when there is only wood charcoal. The ignition and the homogeneous combustion burn the same fuel, pyrolysis gases, when the heterogeneous combustion burns another fuel, wood charcoal. Then, it implied that the emissions of the ignition and homogeneous combustion are comparable. Indeed, their CH_4 ratios (CH_4/VOC) are similar (about 32%) while it is around 69% for the heterogeneous combustion. Finally, the ignition can be considered as a non-optimized combustion, contrary to the homogeneous step.

Table 1. Results on gaseous emissions

	Wood consumption (%)	O_2 (%)	CO (mg.Nm^{-3} at 13% O_2)	COVT (mg.Nm^{-3} Eq CH_4 at 13% O_2)	CH_4 (mg.Nm^{-3} at 13% O_2)	Smoke Temp. (°C)
Ignition	27.7	8	4161	209	66	297
Homogeneous Combustion	56.3	8.9	1046	34	11	285
Heterogeneous Combustion	16.0	14	5509	280	193	227

Concerning the particles emissions, the results are relevant. In the three tests, the EC was heavily, even predominantly emitted during the ignition. The ignition emits a high quantity in mass of TSP but a medium quantity of PM_{10} in number. The homogeneous and the heterogeneous steps emit only few TSP in mass. Nevertheless, the homogeneous combustion is the more emissive step for PM_{10} in number when the heterogeneous is the less emissive. Then, the emissions of particles, whatever the unity is (mass or number), do not only depend on the concentration of CO or VOC in the smoke. The TC ratio (TC/TSP) was about 24% for the ignition, and between 70 and 80% for the other step.

Table 2. Results on particles emissions

	TSP (mg.Nm^{-3} at 13% O_2)	TC (mg.Nm^{-3} at 13% O_2)	EC (mg.Nm^{-3} at 13% O_2)	OC (mg.Nm^{-3} at 13% O_2)	Number PM<10µm
Ignition	50.3	12	3	10	4.E+13
Homogeneous Combustion	2.7	2	0	2	7.E+13
Heterogeneous Combustion	1.3	1	0	1	2.E+13

3.2 Results with EDS and SEM-FEG acquisition

Concerning the ignition, the results of the microanalysis (76% of C in mass) were not consistent with those of the TC ratio described before (24% of C in mass). This disagreement only happened during the ignition. A various sampling period may explain this point. Indeed, the sampling for the TC emission was hold during the entire stage whereas the one for the microanalysis was hold only between the third and the fourth minutes, 30 seconds after that the flame appears. Then, a huge emission of non-carbonaceous particles might occur before or after this sampling time. This emission could be due to two matter points:

- the loading of the logs led to a high emission of flying ashes;
- the non-carbonaceous material contained in a log is quickly extracted of the log and emitted.

The first solution is not possible in this case because every sampling began when the door of the stove was closed. Then, even is the loading emitted some flying ashes, it was

not sampled on the quartz filter using for the TC analysis. Moreover, the results of microanalysis showed that there were more particles with non-carbonaceous materials emitted during ignition (minimum 10% in number) than the other step (less than 2% in number). The explanation could be that the sampling was realized after or before the peak.

The ignition apparently emitted two different kinds of particles with a specific composition (Fig. 1). The Fig. 2 presents two pictures of typical particles emitted during ignition: pieces of wood more or less degraded; nanoparticles more or less aggregated. Then, the both specific compositions might be linked to these both kinds of particles.

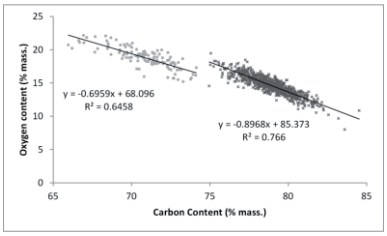

Fig. 1. Carbon and Oxygen composition analysed with EDS of particles from the ignition

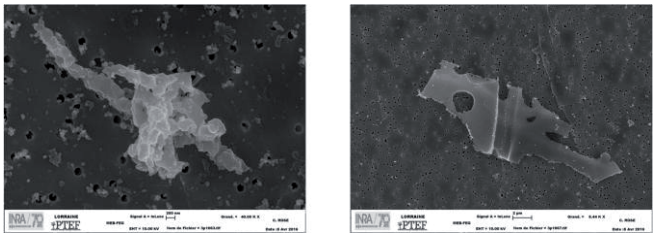

Fig. 2. Pictures (SEM-FEG acquisitions) of particles emitted during the ignition

Concerning the homogeneous combustion, only one group of particles is defined. Indeed, all the particles analyzed have a similar evolution of their C content in function of their O content (Fig. 3). Fig. 4 confirms that only nanoparticles with a diameter around 40 nm, somewhat or no aggregated, were emitted. Schneider et al. (2006) and Tissari et al. (2008) already took some pictures of similar nanoparticles. Moreover, the equation of correlation between the carbon and oxygen content is the same than one group of particles emitted by the ignition. Then, the hypothesis concerning the two different group of particles emitted during the ignition is verified.

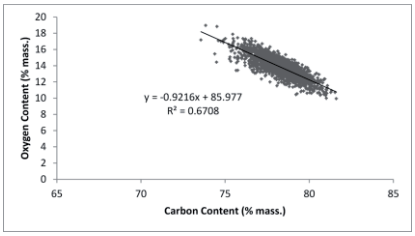

Fig. 3. Carbon and Oxygen composition analysed with EDS of particles from the homogeneous combustion

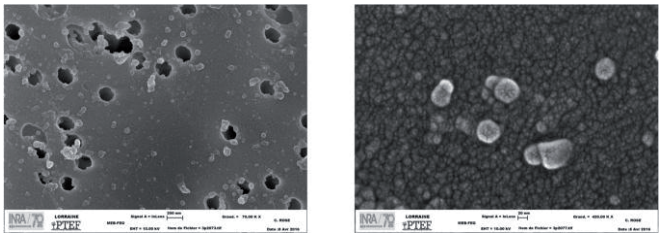

Fig. 4. Pictures (SEM-FEG acquisitions) of particles emitted during the homogeneous combustion

The results for the heterogeneous combustion are totally different from those for the other step of combustion. Indeed, the equation of correlation defined by the evolution of the carbon and oxygen content of the particles is totally different (Fig. 5). The C content average is also the most important (about 80% in mass with the Pt coating). Fig. 6 presents two pictures of the typical particles emitted during this last step of combustion. These particles seemed to be a kind of flying wood charcoal. The results are consistent because the remaining fuel was wood charcoal. Like the stove is designed to burn pyrolysis gases, the air supplies were not efficient during this step and led to a decreased of the temperature. Then the flying wood charcoal could not be burned and is emitted in the smoke.

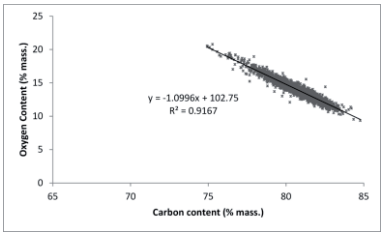

Fig. 5. Carbon and Oxygen composition analysed with EDS of particles from the heterogeneous combustion

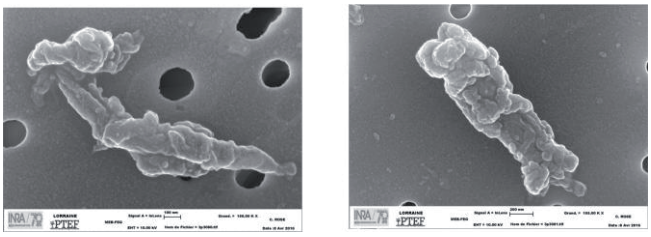

Fig. 6. Pictures (SEM-FEG acquisitions) of particles emitted during the heterogeneous combustion

4. Conclusions

The characterisations of the gaseous and particles emissions for each step of log combustion were achieved. Three main conclusions can be made:

- the ignition emits a lot of pollutants and could be defined as a non-optimized homogeneous combustion because they present the same characteristics (O_2 rate about 8.5%, CH_4 ratio about 30%, Nano-particles…);
- the heterogeneous combustion is responsible for a high proportion of the gaseous emissions but does not emit a lot of particles;

- the SEM-FEG coupled with EDS analyser allowed to characterize the particles composition with a high precision. These processes could be used in order to better understand the formation mechanisms of the particles.

In this performant log stove, four main groups of particles are emitted:

- nanoparticles with a diameter comprised between 30 and 50 nm: ignition and homogeneous combustion;
- piece of wood more or less degraded: ignition;
- flying wood charcoal: heterogeneous combustion;
- a huge quantity of non-carbonaceous material non-totally defined: ignition.

Three solutions are worth considering in order reducing the pollutant emissions:

- optimizing the combustion during the ignition and heterogeneous stage;
- reducing the ignition and the heterogeneous stage time;
- reducing the composition of non-carbonaceous material of the fuel.

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