



## Research paper

## Impact of ignition technique on total emissions of a firewood stove

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## ABSTRACT

A comparison of the effects of traditional stove ignition with paper under the wood versus the Top-Down ignition of a wood crib is made, comparing the gaseous and particulate emissions. Top-Down ignition reduced the unburnt gas emissions by a factor of 2. The Total Suspended Particle (TSP) emission was reduced by about 1/3, while Organic Carbon (OC) was reduced by 73% relative to traditional under-fire ignition. Never the less the Elemental Carbon (EC) doubled in Top-Down ignition. The particle formation mechanism is also different with primary emissions being nanoparticles (20 nm - 50 nm) which agglomerated as they passed along the flue duct. The TSP were generally composed of CHO while the smaller size range < PM<sub>1</sub> and especially the PM<sub>0.1</sub> contained other elements.

## 1. Introduction

Most developed countries depend on fuel, which leads them to increasingly use and improve renewable energies. Saidur et al. [1] demonstrated that domestic wood heating, which is composed of boilers (central heating, hot water) and biomass room heating appliances (room heating, hot air), is the first renewable energy and is the one that is promoted. Moreover, Kalt and Kranzl [2] showed that domestic wood heating is one of the cheapest energies, especially for logs. In this context, the sales of wood domestic appliances have considerably increased in all Europe [3]. In Europe, the residential sector (except for pellets) is the main share of wood energy with 27% of the total use [4]. Then, in this study, the performance of a firewood stove was focused on. Many scientific papers describe this energy as an important source of particles emissions [5–8] particularly during winter [9]. Particles in the ambient air represent an important risk for health because of their size, their surface and their composition [10–14]. Moreover, the composition in Elemental Carbon (EC) and Organic Carbon (OC) of particles induces a modification of the radiative balance of the earth and also changes the composition of the clouds [15–17]. The particles in the atmosphere from wood fuel combustion are not only due to the direct particulate emission but also to a physical and chemical condensation process that produces additional particles from volatile compounds in the flue gases [18]. The Secondary Organic Aerosols (SOA) are an important fraction of the particles in the ambient air [8]. Therefore, the Volatile Organic Compounds (VOC) and the Total Suspended Particles (TSP) need to be measured simultaneously [19]. Even if some

secondary emission abatement systems do exist and are suitable for firewood stoves [20,21], only few installations are fitted. Then the emissions in firewood stoves must first be reduced by the user. Many primary measures were developed, (secondary air, insulation of the combustion room ...) and led to cleaner combustion. Many parameters independent of the user impact the emissions and the thermal efficiency (i.e. chimney system and weather conditions). One of the parameters that most impacts on the emissions is the operating practice [20,22] like fuel characteristics [23], combustion air supply settings [24] or the ignition technique. These improvements of firewood stoves are sustained by the evolution of standards and labels [21]. Nevertheless, the fuel feed and the ignition of firewood stoves remain highly emissive [25]. Indeed, during the ignition phase, the furnace and hearth are cold and lead to high emissions of unburned gas and particles. In order to solve this issue, the Top-Down ignition was recommended in Switzerland. The Top-Down offers an advantage to induce a more progressive ignition. Indeed, as the flame is on the top, the radiation will be more gradual and then the emissions of pyrolysis gases will also be progressive. On the contrary, with a Traditional Ignition system, the whole fuel is directly heated by the combustion. Miljevic et al. [26] demonstrated that this kind of ignition reduced the Total Suspended Particles (TSP) emissions contrary to Vicente et al. [27].

Several different pollutants were followed, namely CO, NO, SO<sub>2</sub>, Total Volatile Organic Compounds (VOC), CH<sub>4</sub>, Total Suspended Particles (TSP), Total Carbon (TC), Elemental Carbon (EC) and Organic Carbon (OC). By measuring the CH<sub>4</sub> emissions separately in addition to the TVOC measurement, it allowed the CH<sub>4</sub>/TVOC ratio to be

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measured. Moreover, so as to obtain a better understanding of the mechanisms of formation of the particles, two microscopes were used.

In order to evaluate the impact of both ignitions modes on a real use of the studied stove, a simulation was also developed.

## 2. Materials and methods

### 2.1. Stove, combustible and experimental procedures

The WABI (D2I INVICTA, DONCHERY, FRANCE) firewood stove that was designed, built and purchased in 2012 with a nominal thermal heat output of 6 kW was used. The combustion chamber was fitted with primary and secondary air supplies and was insulated with vermiculite. The fuel was split beech (*Fagus sylvatica*) logs of 12 cm of diameter from a 40-year-old beech. The logs were chosen without any knots and bark. They were air-seasoned in greenhouses, and their moisture content was stabilized in a regulated enclosed chamber. Then, the water mass fraction was 0.12 (following the standard EN 14774). The wet base Net Calorific Value (NCV<sub>wb</sub>), measured following the standard EN 14918, was 16.7 MJ kg<sup>-1</sup>. The ash mass fraction of the dried material was measured following the standard EN 14775 at 823.15 K and 1088.15 K. The results are: 2.7 g kg<sup>-1</sup> at 823.15 K; 1.4 g kg<sup>-1</sup> at 1088.15 K. The elemental composition of the logs was also measured. In order to obtain a representative result, a complete log was ground up. The sawdust was then mixed, and three samples of 1 g each were analysed. The results were equivalent for each of the three analyses, and are: 495 g kg<sup>-1</sup> of C; 59 g kg<sup>-1</sup> of H; 438 g kg<sup>-1</sup> of O; < 3 g kg<sup>-1</sup> of N; 108 mg kg<sup>-1</sup> of S; 64 mg kg<sup>-1</sup> of Cl.

Two kinds of ignition modes were studied: Top-Down ignition and Traditional ignition (Fig. 1). In both cases, only two sheets of a classic newspaper were used to ignite so as to be close to real conditions. The newspaper used was a paper of 52 g m<sup>-2</sup>, from thermomechanical pulp (made out of 100% chipped conifer). There was only black lead-free ink on this newspaper. Three tests were realised for each configuration. The combustion platform conforms to the EN 13229 and the Pr EN 16510. The fuel was weighed in order to obtain a comparable mass and improve the repeatability. The description of the loads of woods is presented in Table 1. The air valves for combustion air supply were totally open and the combustion was held with a normalized draft (EN 13229; average for every test: 12 Pa; variation: 2 Pa max during a test). In order to evaluate only the ignition period of the stove, the sampling period began at the closing of the fire door and was stopped when the flames disappeared. Normally a standard user would add logs when once flames disappear to keep the fire burning.

### 2.2. Sampling and analysis

#### 2.2.1. Physical and gaseous measurement

During each test, many samplings were realised. The gaseous composition of the smoke was studied thanks to a gas analyser PG 350 (HORIBA, KYOTO, JAPAN) which uses different analysis methods:

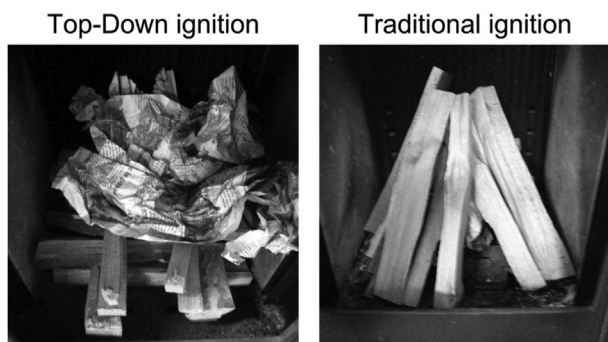


Fig. 1. Top-Down and Traditional ignition.

Table 1

Detailed mass for each load.

Test	Layer 1 (g) <sup>a</sup>	Layer 2 (g) <sup>a</sup>	Layer 3 (g) <sup>a</sup>	Layer 4 (g) <sup>a</sup>	Layer 5 (g) <sup>a</sup>	Layer 6 (g) <sup>a</sup>	Layer 7 (g) <sup>a</sup>	Total (g)
Top-Down 1	418.8	348.7	117.8	116.8	97.7	82.5	57.2	1239.5
Top-Down 2	399.8	335.5	147.1	109.7	92.2	77.2	68.7	1230.2
Top-Down 3	524.1	246.2	156.5	128	96.2	49.7	33.1	1233.8

	Large wood (g) <sup>b</sup>	Medium wood (g) <sup>c</sup>	Kindling wood (g) <sup>d</sup>	Total (g)
Traditional 1	777.4	123.5	258	1158.9
Traditional 2	710.7	240.5	272	1223.2
Traditional 3	633.6	284.7	316.6	1234.9

<sup>a</sup> The layers match the mass of wood at each level from the bot (1) to the top (7) of the load. Each layer is composed by two firewood pieces.

<sup>b</sup> Large wood matches the mass of a group of logs which have a maximal diameter of 4 cm.

<sup>c</sup> Medium wood matches the mass of a group of logs which have a maximal diameter of 2 cm.

<sup>d</sup> Kindling wood matches the mass of a group of logs which have a maximal diameter of 1 cm.

chemiluminescence detectors (NO); Nondispersive infrared sensor (SO<sub>2</sub>, CO, CO<sub>2</sub>); paramagnetic detector (O<sub>2</sub>). The Total Volatile Organic Compounds (TVOC) were measured with a Flame Ionization Detector (FID) Graphite 52M (ENVIRONNEMENT SA, POISSY, FRANCE). A balance with a resolution of 50 g allowed the evolution of the mass of the stove to be followed. The ambient and the flue gas temperatures (at the sampling point, according to EN 13229) were also measured with K-type thermocouples.

#### 2.2.2. Particulate matter sampling and analysis

The Total Suspended Particles (TSP) were out-stack sampled during the entire period thanks to an isokinetic probe at 283 cm<sup>3</sup> s<sup>-1</sup> STP on a quartz filter heated at 160 °C (STP in this study: 0 °C, 101325 Pa). Before and after the sampling, the quartz filter was conditioned according to the Pr EN 16510 (4 h in a heat chambers at 180 °C, 4 h in a desiccator, weight of m<sub>0</sub>, sampling, 4 h in a heat chambers at 180 °C, 4 h in a desiccator, weight of m<sub>1</sub>). For the particles size determination concentration, an Electric Low Pressure Impactor (ELPI) heated at 160 °C was used (DEKATI, KANGASALA, FINLAND). The emissions of Total Carbon (TC), Organic Carbon (OC) and Elemental Carbon (EC) were also measured with a thermal-optical analyser (SUNSET LABORATORY, TIGARD, USA). The sampling was realized at 160 °C on a quartz filter for 100 min, but with a lower flow than the TSP sampling (167 cm<sup>3</sup> s<sup>-1</sup>). The analysis protocol is described deeply by Brandelet et al. [28].

#### 2.2.3. Microscopy for imagery

In order to improve the physical knowledge of the particles, a Field Emission Gun Scanning Electron Microscope (FEG SEM) was used (ZEISS, OBERKOCHE, GERMANY). The filter was in polycarbonate with a porosity of 200 nm. The sampling characteristics were: 16.7 cm<sup>3</sup> s<sup>-1</sup> STP for 25 s at 443.15 K. A secondary electron detector (in lens) was used to obtain high resolution pictures after platinum coating of the filter.

#### 2.2.4. X-ray micro-analysis (EDS)

Many micro-analyses were performed on the same filter as the one observed with high resolution FEG. Automated detection and qualitative X-ray micro-analysis (EDS elementary analysis) of the particles were performed with INCA-Feature module (OXFORD INSTRUMENTS, ABINGDON, UK). The detection of particles was performed from backscattered electron emission by particles (BSD) submitted to 20 kV of acceleration beam voltage in High Vacuum (HV) mode (platinum

coating). 2000 particles were automatically analysed per filter in order to improve the robustness of the results. To avoid spatial segregations, these particles were analysed in two groups geometrically separated on the filter.

### 2.3. Simulation

In order to evaluate the impact of the Top-Down ignition on a heating period, a “conventional life rhythm” was defined. It consists of two heating periods with the objective of checking if the ignition mode could change substantially the emissions of any heating period, including ignition. The first one corresponded to the use of the stove during weekday evenings: the users ignited the stove at 18:00 and stopped heating at 23:00. The second period simulated a weekend with a heating period between 09:00 to 21:00. For these two periods, the first half hour corresponded to the ignition. These heating period, validated by the environment and energy French agency (ADEME), is the one described by the CEREN [29] in 2009 and confirmed by Autret et al. [30] in 2010.

The actual duration for lighting the stove was 26.7 min and 26.5 min for Top-Down and Traditional ignition, respectively. The scenario was calculated with the emissions factors of the steady state and of both ignition modes. The emissions factors of the Traditional and Top-Down ignition modes were determined by six experiments detailed in this study. The emissions factors of the steady state were measured by five additional experiments. For the additional experiments, the air valves for combustion air supply were reduced by 2 in order to follow the settings of the manufacturer.

## 3. Results and discussions

### 3.1. Impacts on gaseous emissions

The Top-Down ignition mode had a high impact on the gaseous emissions. Table 2 presents the results. The O<sub>2</sub> volume fraction of dry gas was around 0.12 and 0.105 for Top-Down and Traditional ignition mode, respectively: indeed, the Traditional ignition mode led to a partial obstruction of the primary air and the loss of mass was more important at the beginning of the combustion. This phenomenon is due to the flame radiation which is more important in the case of Traditional ignition and more progressive in the case of Top-Down ignition.

The duration of ignition looks similar whatever the ignition mode. Then, the duration of ignition, whatever the mode, was estimated at

1800 s (30 min) for the simulation.

### 3.2. Impacts on particles emissions

Concerning the particles emissions, the Top-Down ignition mode was also more efficient (Table 3). Indeed, the TSP emissions were reduced in mass by 32%. Miljevic et al. [26] found a decrease comprised between factors 2 to 5. The decrease is then larger for gaseous emissions than for PM. Nevertheless, thanks to the particle size distribution measurement, it appears that, in both cases, the particles with an aerodiameter higher than 1 µm were a minor fraction (content < 0.001). For the Top-Down ignition, the contents in PM<sub>0.1</sub> and PM<sub>0.1-1</sub> were 80% and 20% in number respectively. In the case of Traditional ignition, the contents were about 70% and 30% in number, respectively. In any case, these results are consistent with the work of Johansson et al. [19] which described the submicronic fraction as the most important. Moreover, Pettersson et al. [31] or Bäfver et al. [32] found the same order of particle size determination.

Many kinds of particles were observed at the SEM and were specific to the ignition mode. First, the Top-Down ignition mainly emitted some agglomerates of nanoparticles (20–50 nm per nanoparticles). Their size could vary, but are generally comprised between 100 and 600 nm (Fig. 2, left). Schneider et al. [33], Tissari et al. [34] and Sippula et al. [23] found some similar aggregates of particles about 20–50 nm of diameter. Regarding the Traditional ignition mode, the most observed particles were a kind of melted material (Fig. 2, right). The main difference between the agglomerate and the melted material was their size (100–600 nm and 0.3–1 µm, respectively). Even if both particles look similar at first sight, they are different. Indeed, in the case of the agglomerate, it is possible to outline each nanoparticle, contrary to the melted material.

The Fig. 3 presents two pictures of typical wood charcoal particles emitted by Top-Down (left) and Traditional (right) ignition. The particles of wood charcoal emitted by Traditional ignition seemed to be less degraded than those from Top-Down ignition. However, the average size was the same (between 3 and 4 µm). Hueglin et al. [35] took similar pictures.

Other kinds of particles were emitted. Fig. 4 shows three pictures of spherical particles bigger than the nanoparticles presented above (0.3–2.5 µm). In fact, these particles were condensate of SVOC, as Chakrabarty et al. [36] supposed. This kind of particles was only observed for Traditional ignition. As Top-Down ignition reduced considerably the TVOC emissions, the same should be applicable for the

**Table 2**  
Top-down and traditional gaseous emissions

Ignition	Repetitions	Mass loss (–)	Test duration (s)	O <sub>2</sub> (Volume fraction) <sup>a</sup>	CO <sub>2</sub> (Volume fraction) <sup>a</sup>	CO (mg m <sup>–3</sup> ) <sup>a,b,c</sup>	NO (mg m <sup>–3</sup> ) <sup>a,b,c</sup>	SO <sub>2</sub> (mg m <sup>–3</sup> ) <sup>a,b,c</sup>	TVOC (mg <sub>CH4</sub> m <sup>–3</sup> ) <sup>a,b,c</sup>	CH <sub>4</sub> (mg m <sup>–3</sup> ) <sup>a,b,c</sup>	Temperature of the flue gas (°C)
Top-Down	1	0.84	1650	0.132	0.074	2131	72	5.7	210	46	241
	2	0.76	1800	0.125	0.081	2266	73	4.6	111	30	231
	3	0.70	1360	0.112	0.092	2183	65	5.4	134	30	249
	<b>Average</b>	<b>0.77</b>	<b>1604</b>	<b>0.123</b>	<b>0.082</b>	<b>2193</b>	<b>70</b>	<b>5</b>	<b>152</b>	<b>35</b>	<b>240</b>
Traditional	1	0.86	1560	0.113	0.090	4893	73	24.6	356	113	254
	2	0.83	1520	0.100	0.102	4151	64	27.1	276	111	270
	3	0.87	1690	0.108	0.095	5044	56	36.8	347	104	271
	<b>Average</b>	<b>0.86</b>	<b>1588</b>	<b>0.107</b>	<b>0.095</b>	<b>4696</b>	<b>64</b>	<b>30</b>	<b>326</b>	<b>109</b>	<b>265</b>

TVOC: Total Volatile Organic Compounds.

<sup>a</sup> measured on dry gas.

<sup>b</sup> STP 0 °C, 1.013E5 Pa.

<sup>c</sup> Corrected at 13% O<sub>2</sub>.

**Table 3**  
Top-Down and Traditional particles emissions.

Ignition	Repetitions	TSP (mg m <sup>-3</sup> ) <sup>a,b,c</sup>	Total particle number PM <sub>10</sub> on wet wood (kg <sup>-1</sup> ) <sup>b,c</sup>	Particles size distribution (number fractions)			
				PM <sub>0.1</sub>	PM <sub>0.1-1.0</sub>	PM <sub>1.0-2.5</sub>	PM <sub>2.5-10</sub>
Top-Down	1	48.8	3.73E+14	0.80	0.20	1.69E-04	6.94E-05
	2	47.4	5.47E+14	0.84	0.16	1.02E-04	4.83E-05
	3	50.1	3.25E+14	0.78	0.22	2.29E-04	9.82E-05
	Average	48.8	4.15E+14	0.81	0.19	1.67E-04	7.20E-05
Traditional	1	77.9	2.66E+14	0.73	0.27	1.46E-04	7.52E-05
	2	65.6	1.01E+14	0.68	0.32	2.16E-04	7.44E-05
	3	72.7	2.25E+14	0.66	0.34	1.58E-04	5.82E-05
	Average	72.1	1.97E+14	0.69	0.31	1.73E-04	6.93E-05

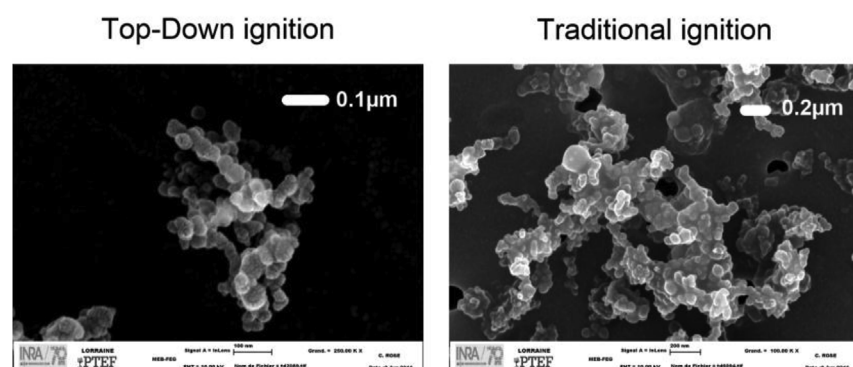
TSP: Total Suspended Particles.

PM: Particulate Matter.

<sup>a</sup> Measured on dry gas.

<sup>b</sup> STP 0 °C, 1.013E<sup>5</sup> Pa.

<sup>c</sup> Corrected at 13% O<sub>2</sub>.



**Fig. 2.** Agglomerate of nanoparticles emitted by Top-Down ignition (\*250000) and melted material emitted by Traditional ignition (\*100000).

#### SVOC emissions.

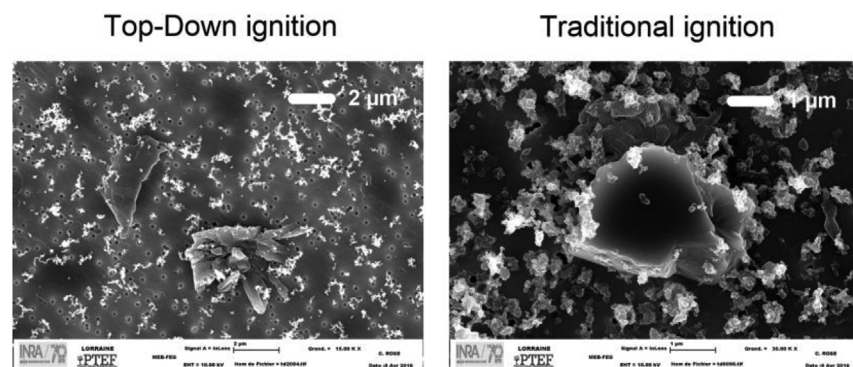
All these particles characteristics led to a specific chemical composition. The OC/EC analyser allows a better characterization of the particles. The OC/EC composition was typical for each kind of ignition mode. The results for the OC/EC analysis are in Table 4. The Traditional ignition mode provides similar EC and OC global ratio than those determined by Schmidl et al. [37].

The composition in carbon was really different and specific for both kinds of ignition modes. All of these OC/EC ratios were consistent with those described in many scientific papers [38,39]. The OC was really less concentrated in the case of Top-Down ignition. Indeed, the OC emissions seemed to follow the TVOC and CO emissions (Figure A1). In

this way, as these emissions were reduced with Top-Down ignition, the emissions of OC were also reduced. Sippula et al. [23] also defined a linear evolution between OC and CO emissions.

However, the TC ratio (TC/TSP in mass) was higher and led to a higher emission of EC. Vicente et al. [27] had already defined the same variation of the TC ratio between both ignition modes.

The micro-analysis produced consistent results. Concerning the elemental composition of the particles, there were two main results. First of all, the ratio of the Oxygen mass fraction and the Carbon mass fraction are linearly correlated. Fig. 5 presents the curves of this phenomenon for Top-Down (left) and Traditional ignition (right). Then, the correlation was the same for the three experiments with Top-Down



**Fig. 3.** Field Emission Gun Scanning Electron Microscope (FEG SEM) pictures of particles of wood charcoal emitted by Top-Down (\*15000) and Traditional (\*30000) ignition.



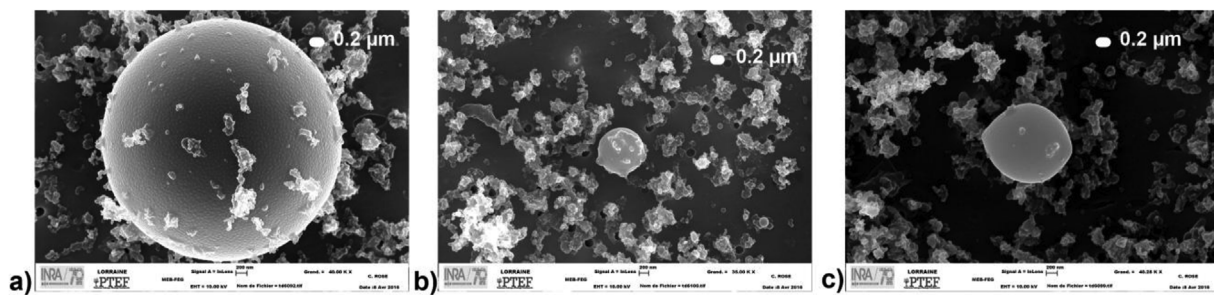


Fig. 4. Field Emission Gun Scanning Electron Microscope (FEG SEM) pictures of spherical particles emitted by traditional ignition (a, \*40000; b, \*35000; c, \*40280).

ignition and a different linear correlation was obtained for the three experiments with Traditional ignition.

Nevertheless, in every test, there was enough oxygen in the smoke to oxidise the particles. Then, the main parameter that could change the composition of the particles could only be the temperature and the mixture of  $O_2$  in the combustion chamber. Indeed, the Top-Down ignition mode induces a more progressive emission of pyrolysis gases by firewood than the Traditional one. This point induces an oxidation also more progressive with the Top-Down ignition mode, and then, a more stable  $O_2$  rate. Indeed, Fig. 6 shows the influence of the temperature of the flue gas on the particles composition: the temperature was the main parameter responsible for the Oxygen and Carbon mass fraction of the particles. Concerning the other elements than Carbon and Oxygen, they were concentrated in the  $PM_{10}$ , and more in the  $PM_{0.1}$ . Fig. 7 represents the evolution of the number of particles containing these elements depending on their size for all the particles analysed for Top-Down ignitions and Traditional ignition. This conclusion was true in both configurations.

### 3.3. Impacts on the emissions scenario

The emissions factors of the steady state, defined by the five additional experiments, are presented in Table A.1. Even if we focus on one evening (Fig. 8), the Traditional ignition mode emitted more pollutants than the Top-Down ignition mode in this firewood stove. The emission of EC was the exception and was reduced by 40% with Traditional ignition. Concerning the NO and the  $SO_2$  emissions, their level of emissions was very low and did not allow observing any consistent

evolution. Then, both pollutants were removed from the total scenario.

The results for one complete heating week followed this trend (Table 5). Then, that meant that if the users of the firewood stove WABI change ignition mode, a consistent reduction of the emissions will occur: the emissions of CO will decrease by 10%; the emissions of TVOC,  $CH_4$ , TSP and TC will decrease by 12%; the emissions of OC will decrease by 40%. Only the emissions of EC are increased by 65% with Top-Down ignition. The total emissions are influenced less than those during ignition.

## 4. Discussions

One hypothesis could explain why the Top-Down ignition mode reduces the emissions of CO, TVOC and  $CH_4$ . Indeed, during Top-Down ignition, the higher average of  $O_2$  volume fraction on the flue gas leads to a better oxidation of the pyrolysis gases. Traditional ignition does not allow burn out of the gas from wood pyrolysis.

Concerning the stability of the emissions of NO, it could be due to the variation of the average temperature of the flue gas (about 20 °C) and of the  $O_2$  volume fraction (about 0.01) which were not sufficient to produce NO by another process than the fuel NO and did not limit or increase the fuel NO production. Then, as the fuel was the same, the NO emissions might be similar. Moreover, Vicente et al. [40] found the same difference of temperature between both ignition modes.

Nevertheless, the emissions of  $SO_2$  were really higher with Traditional ignition. But, the TC mass ratio is really lower in the case of Traditional ignition. This point suggests that the emissions of non-carbonaceous particles were higher with Traditional ignition. Classically, S

Table 4  
Top-Down and Traditional particles carbon compositions.

Ignition	Repetitions	Mass ratio OC/TC	Mass ratio TC/TSP	Global mass ratio OC/TSP	TC ( $mg\ m^{-3}$ ) <sup>a,b,c</sup>	OC ( $mg\ m^{-3}$ ) <sup>a,b,c</sup>	EC ( $mg\ m^{-3}$ ) <sup>a,b,c</sup>
Top-Down	1	0.274	0.959	0.263	46.8	12.8	34
	2	0.198	0.966	0.191	45.8	9.1	36.8
	3	0.121	0.937	0.113	46.9	5.7	41.3
	Average	0.198	0.954	0.189	46.5	9.2	37.3
Traditional	1	0.599	0.774	0.464	60.3	36.1	24.2
	2	0.682	0.645	0.44	42.3	28.9	13.4
	3	0.645	0.776	0.501	56.4	36.4	20
	Average	0.642	0.732	0.468	53	33.8	19.2

OC: Organic Carbon.

EC: Elemental Carbon.

TC: Total Carbon.

TSP: Total Suspended Particles.

<sup>a</sup> Measured on dry gas.

<sup>b</sup> STP 0 °C, 1.013E5 Pa.

<sup>c</sup> Corrected at 13%  $O_2$ .

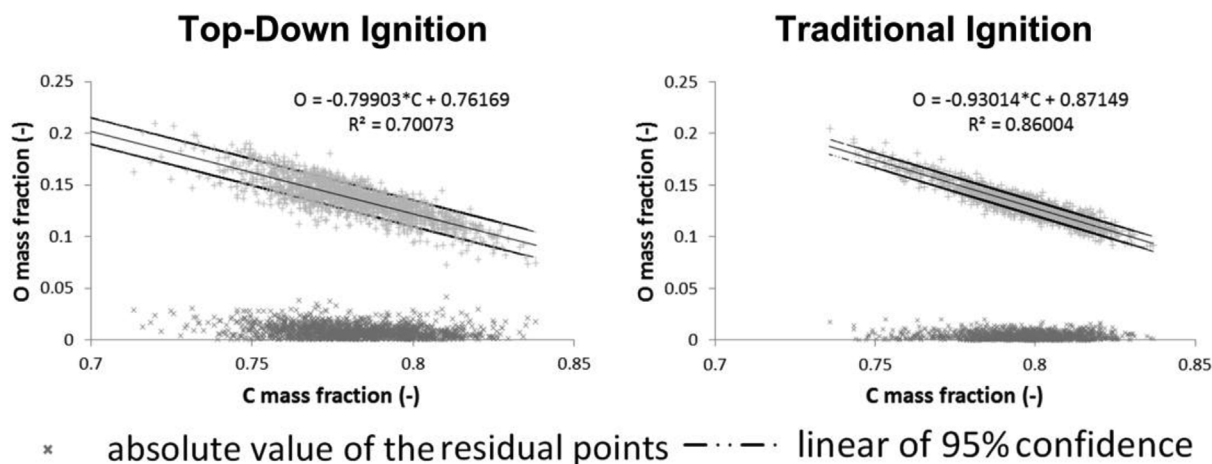


Fig. 5. Correlation between the oxygen mass fraction and the carbon mass fraction of the particles emitted by the third test with both ignition modes.

is in the ash but in the case of Traditional ignition, there were more suspended ashes. The hypothesis of higher emissions of  $\text{SO}_2$  could be that the S in the suspended particles was oxidized and produced  $\text{SO}_2$ .

Two hypotheses of the formation of the nanoparticles could also be discussed. The first one assesses that combustion produced nanoparticles, about 50 nm. Nevertheless, these particles produced some agglomerates which could vary in size. Then, the physical and chemical characteristics of the flue gas were more conducive to the coagulation of the particles in the case of Traditional ignition. This statement was potentially true because of the higher temperature of the flue gas. The particles from Top-Down ignition (Fig. 2, left) could correspond to this mechanism because the nanoparticles were really noticeable. However, in the case of Traditional ignition (Fig. 2, right), the particles seemed to be a melted material. This led to the second hypothesis. The particles were originally some pieces of wood or wood charcoal, and their degradation in the flue gas was like a separation of the material in nanoparticles. In that case, we could come to the conclusion that Top-Down ignition led to a higher combustion rate or temperature than Traditional ignition. In fact, the mechanism of formation of the particles was surely a combination of these two hypotheses. Some nanoparticles were emitted and then agglomerated and some others resulting from a degradation of the wood could agglomerate further in the duct.

In order to determine if the second statement (described above) was

prevailing, the evolution of the emissions of EC could be represented according to the emissions of  $\text{PM}_{10}$  in number (Figure A.2). Indeed, if the number of particles emitted was higher, the second hypothesis would induce that the concentration of organic compounds in particles would be lower because of a better combustion. Then, the EC would be more concentrated. Thus, this slight trend confirmed that the formation of the particles was a combination of two different processes. Moreover, this curve confirmed that nanoparticles were mainly composed of EC. The mass ratio EC/OC depending on the emissions of TSP was drawn in Figure A.3. If the TSP emission was low, the mass ratio EC/OC would have a slight increase. On the other hand, if the TSP emission was low, it meant that the combustion was good and produced many nanoparticles, as the Top-Down ignition. Then, when the particle size distribution was fine the EC mass ratio was higher.

In terms of particle size determination, the results of the microanalysis could be compared to those of the ELPI (Figure A.4). The difference between these both methods was higher in the case of the Top-Down ignition than with the Traditional one. This statement could be explained by the porosity of the filter (200 nm) which induced that the  $\text{PM}_{0.1}$  (which could go through the filter) was under-estimated. However, in the case of Traditional ignition, the results were similar even for the  $\text{PM}_{0.1}$ . The most probable hypothesis was that the ELPI sampling implied a high turbulence due to the high flow and dilution. Then, the

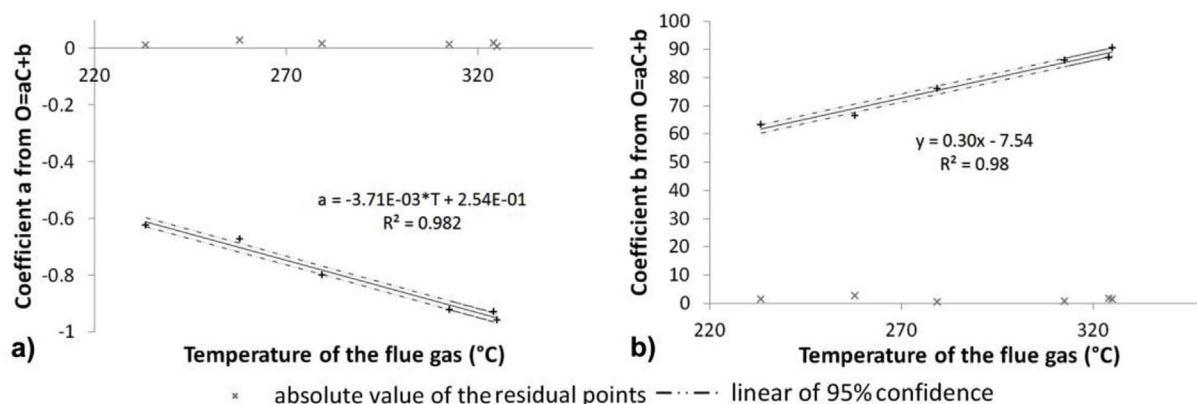


Fig. 6. Correlation between coefficients "a" (a) and "b" (b) from  $O = a \cdot C + b$  with the temperature of the flue gas during the sampling (O is the mass fraction of the particles in Oxygen and C in Carbon).

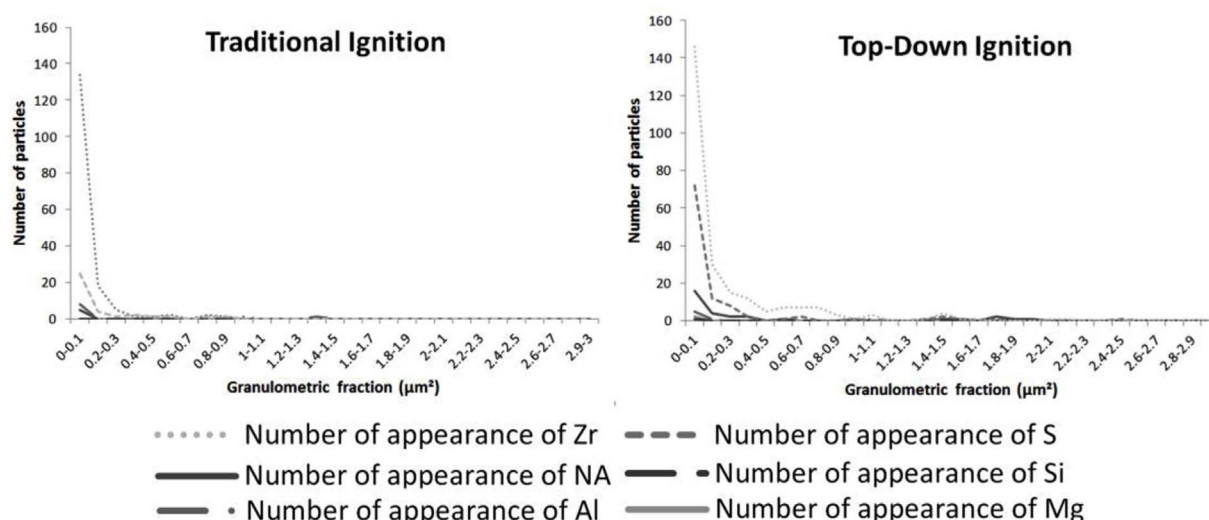


Fig. 7. Granulometric repartition of the particles which contain atomic element different than O and C for Traditional (left) and Top-Down ignition (right).

particles could be destroyed and separated in finer particles. This could not happen in the case of Traditional ignition because the particles were stronger (due to the lower degradation). Moreover, the  $PM_{0.1}$  fraction was more represented in the case of Top-Down ignition. Then, a bigger part of these particles could go through the filter.

The emissions of EC were 100 times higher during ignition than during the steady state. In this firewood stove, the Top-Down ignition mode emits more EC than for Traditional ignition. Then, concerning the emissions of EC, the scenario is not favourable because the ignition mode impacts significantly the emissions of EC. Moreover, Maier et al. [41] showed that PAH emissions (especially benzo(a)pyrene) correlate with EC emissions in boilers. Additional studies on different stoves (including the PAH emissions measurement) could validate whether a generalisation of the Top-Down ignition mode could lead to a high reduction of a majority of the main pollutants. Reichert et al. [42] showed in an investigation that about 8 users out of 10 practice Traditional ignition. Then, if the Top-Down ignition mode is definitely considered more favourable than Traditional ignition, an important communication campaign could change the heating habits of users and could considerably reduce the emissions of domestic firewood stove with a non-technological optimization.

## 5. Conclusion

The use of a firewood stove, and particularly the ignition mode,

considerably impacts emissions. Even at the scale of one heating week, most emissions can be reduced thanks to the Top-Down ignition mode in the studied stove. Only one measured pollutant presents higher emissions with Top-Down ignition: the Elemental Carbon. In order to generalize or not these results, some similar studies must be conducted on other firewood stoves. The PAH emissions have not been measured in this study. Their emissions could follow those of TVOC, and then be reduced by Top-Down ignition. However, some studies have demonstrated that the PAH emissions follow the emissions of EC in boilers. This particular issue needs to be clarified. These two important questions have to be solved before giving any guidance to users.

Nevertheless, the ignition of the firewood stove is still the least efficient step, and therefore the most emissive. As the emissions during the steady state are now really low, manufacturers have to work on the reduction of the emissions during the ignition of the stove.

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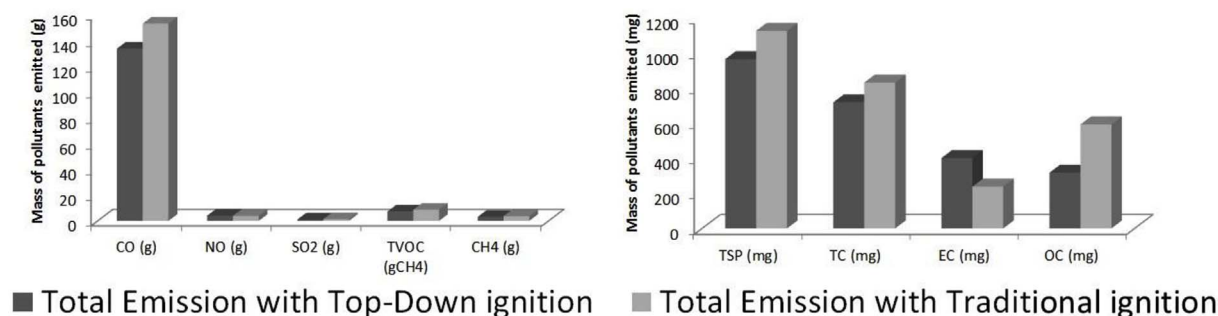


Fig. 8. Emissions of CO, NO,  $SO_2$ , Total Volatile Organic Compounds (TVOC),  $CH_4$ , Total Suspended Particles (TSP), Total Carbon (TC), Elemental Carbon (EC) and Organic Carbon (OC) of one evening following the first period of the scenario.

**Table 5**

Results for the scenario.

	CO (g)	TVOC as CH <sub>4</sub> (g)	CH <sub>4</sub> (g)	TSP (mg)	TC (mg)	EC (mg)	OC (mg)
Total Emission with Top-Down ignition	1295	70	30	8365	5824	2896	2923
Total Emission with Traditional ignition	1432	79	34	9497	6613	1759	4848
Gain (–)	0.1	0.12	0.12	0.12	0.12	–0.65	0.4

Gain = 1-(Top-Down emissions/Traditional emissions).

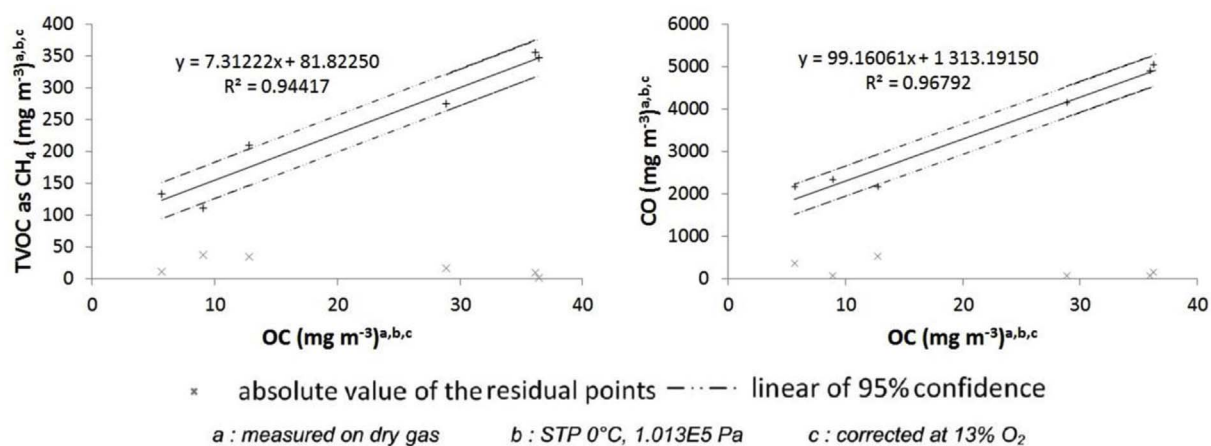
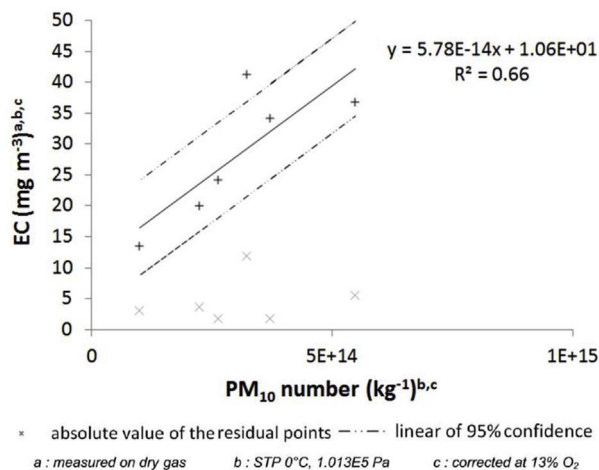
TVOC: Total Volatile Organic Compounds.

TSP: Total Suspended Particles.

TC: Total Carbon.

EC: Elemental Carbon.

OC: Organic Carbon.

**Appendix A****Fig. A.1.** Simultaneous evolution of the emissions of Total Volatile Organic Compound (TVOC) (a) and Carbon Monoxide (CO) (b) with the emissions of Organic Carbon (OC).**Fig. A.2.** Simultaneous evolutions of the emissions of Particulate Matter below 10 µm (PM<sub>10</sub>) and Elemental Carbon (EC).



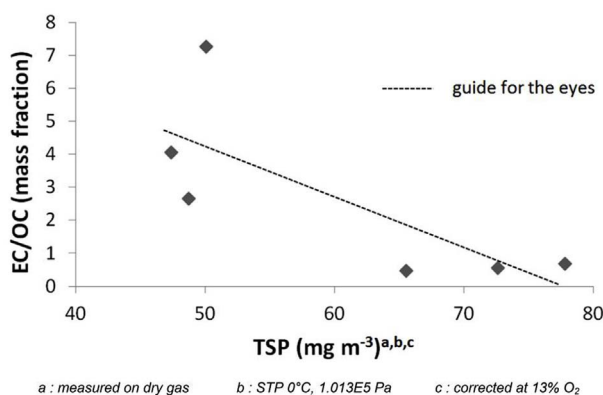


Fig. A.3. Evolution of the ratio between the Elemental Carbon (EC) and the Organic Carbon (OC) composition in mass of the particles with the Total Suspended Particles (TSP) emission in mass.

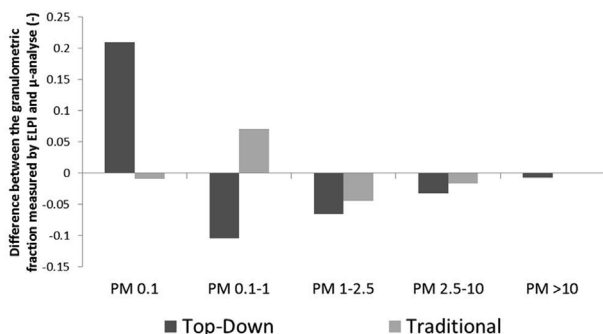


Fig. A.4. Comparison between the granulometric fraction determined by micro-analysis or Electric Low Pressure Impactor (ELPI).

Table A.1

Emissions factors for each configuration used for the scenario.

Configurations	CO	NO	SO <sub>2</sub>	TVOC as CH <sub>4</sub>	CH <sub>4</sub>	TSP	TC	EC	OC
	(g kg <sup>-1</sup> )	(g kg <sup>-1</sup> )	(g kg <sup>-1</sup> )	(g kg <sup>-1</sup> )	(g kg <sup>-1</sup> )	(mg kg <sup>-1</sup> )	(mg kg <sup>-1</sup> )	(mg kg <sup>-1</sup> )	(mg kg <sup>-1</sup> )
Top-Down	21.192	0.679	0.05	1.466	0.346	471	489	392	97
Traditional	38.323	0.526	0.24	2.664	0.89	588	557	202	355
Steady state	15.194	0.45	0.131	0.791	0.366	69	34	3.92	30

TVOC: Total Volatile Organic Compounds.

TSP: Total Suspended Particles.

TC: Total Carbon.

EC: Elemental Carbon.

OC: Organic Carbon.

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