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INFLUENCE OF OPERATING CONDITIONS ON EMISSIONS OF PARTICULATE MATTER FROM A PORTUGUESE WOODSTOVE

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ABSTRACT: Wood combustion experiments were carried out to determine the effect of ignition technique, biomass load and cleavage on emissions from a traditional stove. Two abundant wood species in the Iberian Peninsula were selected: pine (*Pinus pinaster*) and beech (*Fagus sylvatica*). The highest CO and total hydrocarbon emission factors (EF) were observed, respectively, for pine and beech, for the highest and lowest fuel loads. The highest PM_{10} EF was recorded for the operation with the lowest load for both woods. It was found that the top ignition can decrease the PM_{10} EF to less than half when compared with the traditional technique from the bottom. Quantitative analysis of the carbonaceous material showed that the particle mass was mainly composed of OC. The OC content of PM_{10} was higher when loading a lower amount of wood, for beech, or when higher loads were fed into the combustion chamber, in the case of pine. The top ignition contributed to substantial mass fractions of EC in PM_{10}. Regarding particle number distributions, the highest and lowest geometric mean diameters (dg) were recorded with low loads. The highest d_g, for both woods, were observed with non-split wood. Regardless of biofuel or condition tested, 99% of the number concentrations encompassed particles below 400 nm.
Keywords: Residential combustion, Operating conditions, PM_{10}, Size distributions, Flue gas.

1 INTRODUCTION

Biomass combustion is the oldest energy source and is widely applied worldwide for heat and power production, as well as for cooking. Nowadays, due to the strong dependence of developed countries on fossil fuels there have been developed environmental policies that encourage the use of biomass as a renewable energy source. Therefore, it is expected that this source of energy will become more important in the next decades due to the declining fossil fuel supply [1,2]. Although representing a CO_2 neutral source of energy, emissions from residential wood combustion have been considered as a great contributor to air pollution. In Portugal, it was estimated that 2 Mton of wood are annually burned in residential wood combustion, which contributes to particulate emissions representing around 30% of the total particulate matter < 10 µm (PM_{10}) emitted by the diverse sectors of activity [3]. Recent studies [4,5,6] confirm the negative health effects related to residential wood combustion emissions. The emissions resulting from incomplete combustion contain hazardous pollutants to human health, such as carbon monoxide (CO), particulate matter (PM) and polycyclic aromatic hydrocarbons (PAHs) [7]. Huang et al. [8] have been following cardiovascular disease patients during two years and observed a reduction in heart rate variability index due to reduced exposure to $PM_{2.5}$ and black carbon. In addition to negative effects on human health, the airborne particles arising from residential combustion cause disturbances in atmospheric chemistry and climate [9,10]. Taking into account the impact of residential wood combustion emissions and the need for compliance with legal norms, a rigorous quantification and characterization of emissions from this sector is necessary in order to make possible to evaluate and apply suitable mitigation measures to old and modern wood-burning appliances.

Previous work suggested that the wood-fuel and type of appliance used in residential wood combustion have great influence on emissions from this sector [3,11]. Although an extensive characterization and quantification of the residential wood combustion emissions have been made in the recent years, there is still a limited understanding of the influence of operating conditions. In most of the real cases, domestic users make an irregular utilization of the combustion appliance inducing different combustion conditions. The aim of this study is to determine differences in emission factors for distinct operating conditions, representative of the typical domestic use by householders. The parameters that had been investigated were the ignition technique (upside down and bottom up lighting), fuel load and, in the case of high load, split and non-split logs.

2 MATERIALS AND METHODS

2.1 Appliance, fuels and experimental procedures

Combustion experiments were conducted in a typical

Portuguese stove, commonly used for domestic heating, manufactured by Solzaima (Sahara model). This equipment is made of stainless steel with a cast iron grate. The combustion chamber has a volume of 0.093 m^3, corresponding to a height of 0.44 m, a width of 0.59 m and a length of 0.36 m. It is a discontinuous feeding device with manual control of combustion air. The grate was connected to a weight sensor in order to enable the quantification of the fuel mass loss during the combustion tests. The flue gas was exhausted through an externally insulated stack, from the bottom of the chimney up to about 1 m above the outlet of the stove for operational safety, of 0.2 m in diameter and 3.96 m tall. The driving force for the air flow rate throughout the combustion chamber is the natural convection resulting from the up flowing stream of combustion hot flue gases in the chimney. The flow rate of combustion air entering the combustion chamber was determined by a mass flow meter (Kurz Model: 500-40 0.0 P-2).

This work is based on experiments carried out with two types of woody biomass, namely pine (*Pinus pinaster*) and beech (*Fagus sylvatica*), two abundant species in the Iberian Peninsula and commonly used in residential fireplaces and stoves. The first species is a softwood, whilst the second is a hardwood. The elemental composition, ash and moisture content of both fuels are presented in Table I.

Table I: Elemental composition (dry basis), ash and moisture content of biofuels (wt.%).

	Pinus pinaster	***Fagus sylvatica***
Moisture	9.9	9.6
Ash	0.40	1.80
C	51.40	47.97
H	6.20	6.26
N	0.16	0.04
S	bdl*	bdl*
O**	41.84	43.93

*below detection limit of 0.01% w/w; **by difference

To investigate the influence of fuel load on emissions three different load conditions were tested. The fuel was inserted into the combustion chamber in batches ranging between 1 and 4 kg (Table II), depending on the load condition under analysis. These experiments were initiated by putting a batch of fuel on a bed of hot charcoal when the temperatures in the combustion chamber were around 100 °C. All the tests performed to evaluate the effect of load and log cleavage (split (S) and non-split (NS) logs) on emissions correspond to "hot start" conditions.

The wood was cut into logs of 30-40 cm in length, whether for split or non-split logs. The duration of each combustion cycle depended on the operating condition and wood type, ranging from 30 to 95 minutes.

The comparison of different ignition techniques included the conventional ignition from the bottom and ignition from the top. To initiate the combustion experiments of ignition from the bottom, two pine-cones were kindled with a lighter and the fuel load was put on the top of the latter. Ignition from the top was achieved using small pieces cut from the same wood being burned and pine-cones cracked on the top of a batch of logs. These experiments were made with batches of 2 kg of wood for both fuels (medium load). The total number of batches for the test of each technique was between three and five.

Table II: Wood combustion tests and fuel batch details.

	Cleavage	**Load**	**Fuel burnt (kg)**	**Number of batches**
Pinus pinaster	Split	Low load	1.0-1.2	5
		Medium load	1.9-2.1	5
		High load	3.7-4.1	3
	Non-split	High load	3.9-4.0	3
Fagus sylvatica	Split	Low load	1.0-1.2	3
		Medium load	1.8-2.1	3
		High load	3.8-3.9	3
	Non-split	High load	3.8-3.9	3

2.2 Flue gas monitoring

The sample to the gas analyzers was taken straight from the stack with a water-cooled sampling probe. The exhaust gas was measured continuously with on-line analyzers for O_2 (paramagnetic analyser, ADC O2-700 model), CO_2 and CO (non-dispersive infrared analyzer from Environnement, MIR 9000 model). For total hydrocarbons (THC), expressed as CH_4, an externally heated probe and sampling line (at 190ºC) were used. Concentrations in the flue gas were obtained by flame ionization detection (Dyna-FID Hydrocarbon Gas Analyser, model SE-310).

2.3 PM measurement and analyses

Before particle measurement, the flue gas was diluted in a dilution tunnel. The flue gas dilution is widely used (e.g. Fernandes et al, 2011; McDonald et al, 2000; Pettersson et al, 2011; Schauer et al, 2001; Fine et al., 2004a, 2004b; Hedberg et al., 2002; Tissari et al., 2008) to simulate the rapid cooling and mixing that occurs when the exhaust gases are released into the atmosphere. Dilution factors used in the dilution tunnel were around 1:25. The volumetric gas flow rate throughout the tunnel, and respective combustion gas dilution ratio, were calculated from the mean gas velocity in the cross section of the dilution tunnel. The mean gas velocity was estimated from the differential pressure monitored by a Pitot tube and respective pressure sensor (Testo AG 808) and a K-type thermocouple [12,13]. The particulate matter (PM_{10}) samples for gravimetric and chemical analyses were collected on 47 mm diameter quartz fibre filters using an Echo PM sampling head (TECORA, model 2.004.01, Italy) operating at a flow of 2.3 m^3 h^{-1}. The temperature in the particle sampling point was in the range 25–35°C, and the relative pressure was typically 10 mm H_2O below the atmospheric pressure. The filters were kept for 48 h in a room with controlled temperature and humidity before gravimetric quantification with a microbalance (RADWAG 5/2Y/F). Before use, quartz fibre filters were baked at 600 ºC for 6 hours to eliminate organic contaminants. Filter weight was obtained from the average of six measurements, when the variations were less than 0.02%. The carbonaceous content (OC and EC) of the particulate matter collected on the quartz filters was analyzed using a thermal optical transmission technique [13].

The particle number concentrations and particle number size distributions are also important parameters to develop a quantitative assessment of their impacts on both human health and the environment. The particle number size distributions were measured with a laser spectrometer (Passive Cavity Aerosol Spectrometer Probe, PMS Model PCASP-X) in a second dilution tunnel with an internal diameter of 0.07 m and 1.56 m length. The device measures the size distribution of particles with nominal optical diameters between 0.1 and 10 μm in 31 discrete channels. To obtain the exact number concentration of particles in each channel it is necessary to made several corrections to the spectrometer counts. Since the optical particle counter was calibrated with latex particles with a refractive index of 1.58–0.0i, it is necessary to make the refractive index correction for the aerosol sampled. The correction of the raw size bins in order to obtain the exact number concentration versus exact size was done using a program based on the Mie Theory. The refractive index is a parameter used to describe the scattering and absorption characteristics of a particle. Different methodologies have been proposed to estimate the particle refractive index [e.g. 15, 16, 17]. Due to the lack of information on optical properties of the sampled particles, the methodology proposed by Levin et al. [17] seemed to be the most suitable, since it was intended to estimate the refractive index of particles from their chemical composition, assuming that the PM_{10} constituents are present in the form of certain chemical compounds, having a specific density and typical refractive index. Since it was found that carbonaceous constituents accounted for the largest particle mass fraction (42-96%), the refractive index was estimated based on these compounds. It was also necessary to make the adjustment of the sample volume according to the altitude of the sampling point. This correction was made as recommended by the manufacturer.

3 RESULTS AND DISCUSSION

3.1 Combustion conditions

For each combustion test, the continuous monitoring of the following parameters was perfomed: 1) fuel load on the grate, 2) combustion air flow rate entering the combustion chamber, 3) temperature of the exhaust gas in the combustion chamber and in the chimney, and 4) O_2, CO_2, CO and THC concentrations.

The combustion cycles were characterized by three main stages, namely ignition, flaming and smouldering. The first stage corresponds to the heating of the biomass followed by drying and initial steps of devolatilization, without the existence of visible flame. This initial stage of combustion is a critical step, since the fast devolatilization of the fuel originates high amounts of CO and THC in a short time. These compounds are not oxidized, despite the high concentration of O_2 available. This can be explained by the low temperatures in the combustion chamber and poor mixing between the combustion air and flue gas. It was observed that the O_2 content of the flue gas starts to decrease while the temperature in the combustion chamber increases. As long as the O_2 concentrations are too high and the temperatures in the combustion chamber are too low to achieve appropriate burnout conditions, high CO and THC concentrations in the flue gas are detected. The flaming stage is characterized by devolatilization and combustion of volatiles and char during which a vigorous flame is observed. In this stage the O_2 availability in the flue gas and sufficiently high temperatures in the combustion chamber provide acceptable burnout conditions. During this phase, the oxygen concentration in the flue gases decrease rapidly, as well as the THC and CO concentrations. On the other hand, the CO_2 concentration in the flue gas increases due to the oxidation of volatiles. At the end of the combustion cycle, char combustion takes place. In the smouldering stage, the O_2 concentrations in the flue gas start to increase and the temperatures in the combustion chamber decrease again. Consequently, the burnout quality decreases and the CO emissions increase.

The maximum CO concentrations for the combustion of pine occurred during the operation with high fuel load using split (7546 ppmv, dry gases) and non-split logs (7661 ppmv, dry gases). The combustion of beech showed the same trend, also with high values during the operation with high loads of both split (8045 ppmv, dry gases) and non-split logs (7411 ppmv, dry gases). The maximum THC concentrations were also recorded under the same operating conditions: 3401 ppmv (wet gases) for pine and 4031 ppmv (wet gases) for beech. The fuel load had a clear effect on emissions. The use of high loads, with and without cleavage, may lead to incomplete combustion due to the insufficient combustion air supply and poor mixing between the combustion air and flue gas.

Higher flow rates were recorded for high load operation and lower flow rates for low load operation. The flow rate ranged between 23 and 35 $Nm^3\ h^{-1}$ for the operation with low loads for both fuels and between 22 and 47 $Nm^3\ h^{-1}$ for the operation with high loads. This observation can be explained by the higher temperatures reached during the operation with high loads, resulting in more air entering the combustion chamber. Regarding the evolution of the combustion air flow admitted to the combustion chamber during a combustion cycle, there is a sudden increase during the first few minutes (5-10 minutes), until it reaches its maximum value and then decreases gradually until the end of the test. There is a direct relationship between the temperature in the combustion chamber and the combustion air flow. This occurs since the air flow admitted into the combustion chamber is induced by the high temperatures therein. Thus, the higher the temperature the greater natural convection induced by the hot gases in the chimney and thus the greater the combustion air flow. During the combustion at high load conditions, the temperatures in the combustion chamber, for pine and beech combustion, reached a maximum of 749 and 785 °C, respectively. Burning under low load originated maximum temperatures between 607 and 499 °C, for pine and beech combustion, respectively.

With respect to biomass consumption, it was possible to distinguish two phases. In the first one, a high fuel consumption rate was observed, while in the second phase a slow loss of mass over time was registered, which corresponds to the smouldering stage of the combustion cycle. For medium load combustion conditions, it seems that the first phase occurred during the first 30 minutes and the second phase from this moment until the end of the combustion cycle. The duration of both phases of fuel consumption are related to the duration of the entire combustion cycle, which in turn is a function of the fuel loading. For the operation with medium loads, the combustion cycles lasted approximately 60 minutes. Contrary to the conditions of medium loads, for which the duration of a cycle is very similar between batches, the combustion cycles with low and high fuel loads showed highly variable durations. This variability results in different fuel conversion rates and thus different durations of the first and second phases of biomass consumption. The variability observed may be related to operational problems arising from the use of too low or high loads. Low fuel loads have contributed to lower combustion temperatures and reduced char beds, causing difficulties on the ignition of the next fuel batch (combustion chamber recharge). Thus, it is generally observed that during the first refuelling after heating the chamber with a medium load of fuel, temperatures were moderate in the chamber, ignition occurred easily, and the cycle presented a short duration (about 30 minutes). In subsequent batches, ignition was delayed, since the bed of hot coal was reduced, and the cycle took twice as much time. However, after performing multiple batches, the temperature rose whether in the combustion chamber or in the char bed. For high fuel loads, the combustion cycles lasted from 40 to 80 minutes. In the combustion of high loads with cleavage, the fuel was very compressed, hindering the diffusion of oxygen through the pieces of wood, which caused a barrier to initiate the fuel ignition. In the tests where no cleavage was applied to the logs, difficulties in lighting the fire were again felt and low fuel consumption rates were observed, which may arise from the limited contact area between the fuel and the flame.

The use of different ignition techniques produced no significant effects whether on the combustion air inlet or on the biomass consumption. Since the load used to start-up the fires is equivalent to those used for evaluating the emissions of medium fuel loads, the air inlet and biomass consumption profiles are quite similar in both cases. However, it is necessary to take into account that the cold start involves lower temperatures and, consequently, lower combustion air flows at the beginning of the combustion cycle. Comparing the average combustion air flow for the hot start-up (medium load) and cold start-up experiments, only slight differences were observed (about 1 Nm^3 h^{-1} higher when initiating hot start-up combustion tests).

3.2 Gaseous emission factors

Figure 1 displays CO and THC emission factors, for pine and beech combustion, obtained for each condition tested. The highest emission factor for pine combustion occurred for high load when using split logs (1739 g kg^{-1} of wood burned, dry basis). For beech, the condition that generated the highest emission factor was the medium load (1634 g kg^{-1} of wood burned, dry basis). The highest CO emission factor (73.9 g kg^{-1} of wood burned, dry basis) for pine occurred when the combustion at high load conditions with non-split logs was carried out. For beech, the condition that produced the highest CO emission factor (72.6 g kg^{-1} of wood burned dry basis) was the low fuel load. Similarly to the CO emission factors, the highest THC emission factor for pine combustion occurred for high load conditions without cleavage (9.0 g kg^{-1} of wood burned, wet gases). For beech combustion, the condition that generated the highest THC emission factor was low load (10.7 g kg^{-1} of wood burned, wet gases). The use of high loads, with and without cleavage of the logs, may lead to incomplete combustion. This occurs because a large fuel batch increases the rate of devolatilization of the fuel, which causes an insufficiency on the combustion air supply [18]. In equipment with no secondary combustion chamber, the load is a critical aspect, since the empty space in the upper part of the chamber acts as a secondary combustion chamber. When using an excessively high fuel load this space is reduced and consequently the residence time is not enough for mixture of the flue gas and combustion air and gas phase oxidation to occur [19]. Amorim et al. [20] assessed, among other variables, the influence of biomass size and combustion air flow rate on gaseous emission factors. The statistical analysis performed revealed that the most influential parameter was the biomass size. The CO_2 emission factors were higher when using biomass with smaller diameter. Conversely, the CO and CH_4 emission factors were higher when combusting with larger log sizes. This trend was also observed in this study. The emission factors presented by Amorim et al. [20] are close to the values obtained in the present study for beech combustion. The lower emissions of incomplete combustion products when using smaller pieces result from the fact that they produce a longer flame combustion phase and a shorter smouldering phase. Hytönen & Jokiniemi [21] conducted tests in a combustion stove with primary and secondary air inlet. The tests evaluated the influence of moisture content (10 to 25%), fuel load (1, 2 and 3 kg), and degree of cleavage. They found that the emissions cannot be reduced, even by modifying the air combustion flow, if a low fuel load is used. In these conditions, the combustion intensity was low, not allowing the mixing of the flue gases and combustion air. At high loads, they observed high combustion rates, making it possible to obtain an adequate mixing degree, although combustion air needs have been high. The highest degree of cleavage (over 30% of surface area) and lower moisture content generated greater CO emissions for high fuel loads. When applying a high degree of cleavage at low loads and increased moisture content, a reduction in emissions was observed. The explanation lies in the fact that when the reaction area is high and the moisture content is very reduced the combustion is more intense and can lead to local conditions of oxygen deficit. The high moisture content controls the combustion rates, producing lower emissions of unburned products.

It can be seen that there is variability in emission factors, in spite of performing a comparison between the same wood types, similar loads and combustion equipment. This variability may derive from several factors. There is variability in the composition of the biomass, even within the same species, which can induce differences in gaseous emission factors. Combustion conditions and operation are also very important factors. For example, high rates of combustion result in increased emissions of unburned products, which can be induced by the low moisture content of the biofuel [22]. The distribution of biomass itself on the grate and degree of compaction can cause significant differences in emissions [23].

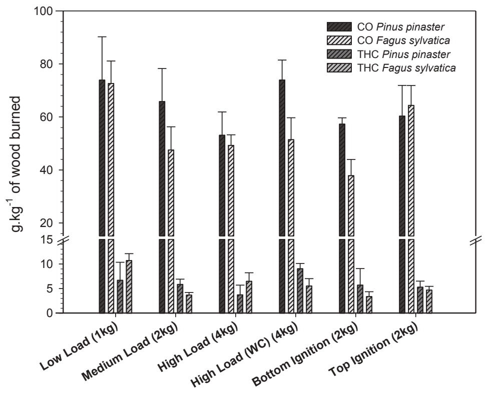

Figure 1: CO emission factors (g kg^{-1} wood burned, dry basis) and THC emission factors (g kg^{-1} wood burned, wet gases)

The CO/CO_2 ratio is a measure of the relative combustion efficiency in terms of conversion of the fuel. A high ratio reflects lower combustion efficiency. The typical ratio for flame combustion phase is less than 0.1 [12]. On average, the ratios obtained for all combustion experiments were below this value, which indicates that the flame combustion phase was dominant. The highest ratios were obtained for the tests with low load operation for both biofuels.

There is a clear relationship between CO and CO_2 emission factors and combustion temperatures. Thus, CO emission factors increase with decreasing combustion temperatures, while the CO_2 emission factors have an opposite behaviour, increasing with increasing combustion temperatures. The technology of combustion is therefore a key factor in the emissions of these compounds. Advanced combustion technologies allow the reduction of CO, enhancing its oxidation to CO_2. On the other hand, fireplaces, where there is a high combustion air inlet reducing combustion temperatures, originate high CO emission factors.

3.3 Particulate emissions and carbonaceous content

The emission factors for "softwood" combustion ranged between 6.50 ± 1.59 and 16.74 ± 1.92 (g kg^{-1} of wood burned, dry basis). The lowest PM_{10} emission factor was observed when operating the stove with medium load, while the highest emission factor was generated when loading a lower amount of wood. The difference found was more than twice, as can be seen in Figure 2. Particulate emissions increase with the decrease of the wood surface area, i.e., when using less cracked wood. The top-down ignition can reduce the PM_{10} emission factor to less than half when comparing with the traditional technique. The traditional ignition technique from the bottom causes the simultaneous firing of the entire batch of fuel, leading to high combustion rates and therefore oxygen deficit zones in the combustion chamber, which results in incomplete combustion. On the other hand, the top-down ignition promotes a gradual combustion of the fuel batch resulting in more complete combustion [24]. PM_{10} emission factors obtained for beech combustion for each operating condition are presented in Figure 2.

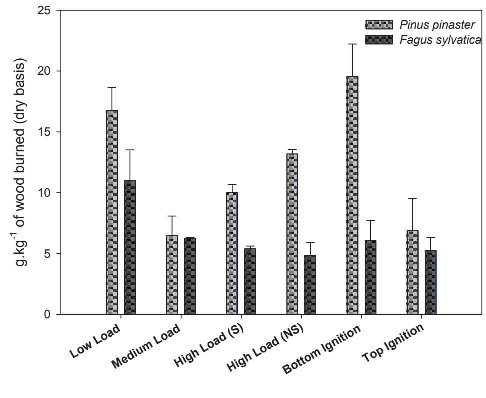

Figure 2: PM_{10} emission factors

The emission factors for the hardwood combustion ranged from 4.86 ± 1.06 to 11.0 ± 2.50 (g kg^{-1} of wood burned, dry basis) for the different loads tested. Contrary to what was observed for the softwood combustion, it seems that, except for low loads, the other operating conditions present similar emission factors. Compared to the bottom lighting, a slight reduction of the emission factor was observed when the ignition of the hardwood was carried out from the top. As observed for the combustion of pine, the highest emission factors were generated under low load conditions. However, contrary to what was registered for the softwood combustion, the lowest emission factor was generated when operating under high loads without cleavage. The lower emission factor may be associated with a higher density of hardwood that promotes lower combustion rates compared to those observed for softwood. Hardwood burns more slowly and, thus, prevents the occurrence of oxygen deficit zones in the combustion chamber. The high temperatures recorded during operation with high loads, associated with good combustion air supply, promote a more complete combustion [23]. Similarly to what was observed in this study for the hardwood tests, Schmidl et al. [25] found that combustion with "high fuel load" in a sophisticated stove presented lower PM_{10} emissions compared with the "normal" operation condition. The authors point out the reduced air combustion flow as a critical factor in particulate matter emissions. Tissari et al. [26] conducted tests on a conventional masonry heater in which two conditions have been tested, namely “normal combustion” and "smouldering combustion". The "smouldering combustion" conditions have been created using a high fuel load and by restricting the entry of combustion air. For normal combustion conditions the authors obtained an emission factor of 1.8 ± 0.5 PM_1 g kg^{-1} of wood burned. For "smouldering combustion" conditions, the emission factor was about six times higher, with a mean value of 11.1 ± 3.9 g kg^{-1}. This value is identical to that obtained in this study under low load condition for beech combustion (11.0 g kg^{-1} of wood burned, dry basis). Thus, reduced combustion temperatures, poor mixture between the flue gas and combustion air or short residence times contributed to increased emissions of particulate matter. The particle emission factors found in the literature may show great variability, due to the fuel characteristics, combustion equipment design, combustion conditions, dilution techniques and sampling procedures [e.g. 19, 26, 27, 28].

3.4 PM_{10} carbonaceous content

There are three main types of primary particles emitted by the combustion process: soot particles, organic and inorganic particles. The distinction between them is based on their formation mechanism and origin. Soot and organic particles originate from combustible material and are formed due to incomplete combustion. The inorganic particles consist of non-combustible material [29] and are formed in conditions of nearly complete combustion [30]. Under incomplete combustion conditions, due to the low combustion temperature and/or local conditions of oxygen deficit, CO and THC significantly increased. In addition to the increased soot formation, organic compounds condense on the surfaces of primary particles or may form very small organic particles through nucleation mechanisms [18].

In manually operated combustion appliances incomplete combustion conditions are dominant. Thus, soot and condensable products are the dominant fraction of the particulate matter released into the atmosphere. Since soot and condensable formation are related to distinct temperatures and residence times, usually only one type of particle is dominant [30]. It was observed that, compared to soot from diesel engines that has been identified as being carcinogenic, particulate matter originated from incomplete combustion has significantly higher concentrations of PAH, as well as much higher levels of toxicity and carcinogenic potential [31].

Table III displays the PM_{10} carbonaceous components for pine and beech combustion, under different operating conditions. Organic and elemental carbon represented 37.0 to 81.8% of the particulate mass emitted during the combustion process. OC was the predominant fraction in all conditions tested. The OC content of PM_{10} ranged between 30.8% (reduced load) and 57.7% (high load). The EC content ranged from 8.2% (high load without cleavage) to 28.9% (medium load). Compared to the bottom lighting, the PM_{10} carbonaceous content was higher when the top-down ignition technique was followed (55.9%). The ignition from the top generated substantially higher fractions of EC (25.4% of PM_{10}) compared with the ignition from the bottom (6.7% of the PM_{10}).

The total carbon content of particles emitted from the beech combustion ranged from 53.8 to 78.0%. The OC content was, as for pine combustion, prevalent in all conditions tested. The PM_{10} OC content ranged from 38.9% (high load) to 58.8% (low load). The EC content ranged from 11.9% (high load without cleavage) to 21.4% (high load). The carbonaceous mass fraction was higher when the top-down ignition technique was applied (74.1%). The top-down ignition generated significantly higher fractions of EC (33.4% of PM_{10}) compared with the traditional technique (23.4% of the PM_{10}). Higher combustion temperatures and vigorous flame conditions potentiate the emissions of soot particles, increasing EC emissions. The increase in the reaction area of the fuel provides these conditions, increasing the EC formation.

Improved combustion efficiency in the combustion chamber, created by higher combustion temperatures and stronger flaming conditions, was probably achieved when the ignition is started from the top. These conditions lead to the formation of more soot particles and therefore enhance the EC emissions. Gonçalves et al. [32] observed a higher EC fraction in particles emitted from a chimney type log woodstove, which was explained by the improved combustion efficiency in the combustion chamber. Gonçalves et al. [33]. evaluated the influence of cold and hot start-up conditions on the carbonaceous content of particulate matter emitted by traditional appliances. The results revealed that the start temperatures of a combustion cycle does not have significant influence on the OC and EC fractions on particulate matter, since the maximum temperatures reached in these appliances do not exceed 600ºC, resulting in the formation of mainly organic particles .

Table III: Carbonaceous components in wood smoke (wt.% PM_{10}).

Biomass type	Operating condition	OC (wt.% of PM_{10} mass)	EC (wt.% of PM_{10} mass)	OC/EC
Pinus pinaster	Low Load	26.8	10.2	2.8
	Medium Load	39.9	22.6	1.6
	High Load (S)	51.3	14.7	2.8
	High Load (NS)	48.2	7.6	6.2
	Top Ignition	35.1	30.8	1.2
	Bottom Ignition	31.5	7.8	4.0
Fagus sylvatica	Low Load	61.8	20.0	3.1
	Medium Load	42.1	19.9	2.1
	High Load (S)	48.0	8.6	1.8
	High Load (NS)	41.4	11.8	3.5
	Top Ignition	41.1	33.9	1.2
	Bottom Ignition	33.9	23.7	1.5

Another observation from the present study is that the wood splitting level caused similar effects on the combustion of both biomass types. High loads with cleavage potentiated the formation of EC (20.7-21.4%), whereas when there is no cleavage applied to the logs, the EC fraction in particulate matter fell substantially (from 8.2 to 11.9%).

The OC/EC ratio can be useful to apportion different sources of carbonaceous particulate matter. Emissions from fossil fuel combustion are characterized by lower ratios, while higher ratios are typical of biomass burning [12, 32, 34]. McDonald et al. [34] obtained an OC/EC ratio of 3.9 for softwood and 7.9 for hardwood in a woodstove. Fernandes et al. [12] reported an average OC/EC ratio of 0.85 for softwood combustion and between 3.14 and 4.39 for hardwood combustion in a chimney type log woodstove. An average ratio of 15.4 ±

7.24 was obtained by Alves et al. [35] for the wood combustion experiments performed in a fireplace and woodstove using Portuguese biomass fuels. In this study, the highest OC/EC ratio was achieved when the bottom-up technique of lighting was applied to pine logs, while the lowest ratio was observed for the same fuel when the fire was ignited from the top. Bølling et al. [36] observed that particulate matter resulting from poor combustion conditions with low temperatures at the combustion chamber in old woodstoves are dominated by OC, corresponding to EC/TC ratios from 0.01 to 0.11. On the other hand, EC/TC ratios for incomplete combustion yet with high temperatures in the combustion chamber that are commonly achieved in stoves and masonry heaters have been reported to range from 0.5 to 0.75 [36].

3.5 Numerical particle concentration

As regards to changes in the total number of particles emitted during a combustion cycle, it was possible to distinguish three phases, according to the observed temperatures in the combustion chamber. Phase 1 corresponds to the heating step, during which it is observed an abrupt increase in temperature. Phase 2 corresponds to the intense burning step, during which the highest temperatures in the combustion chamber are recorded. The third stage corresponds to the flameless combustion stage, during which there is a decrease in temperature in the combustion chamber. These phases, therefore, correspond to the stages of the combustion process. It was possible to observe that the numerical particle concentrations are higher during the flaming phase and smaller during the heating and smouldering stages.

The particle formation starts with the generation of condensation nuclei in the flame zone, in which the majority of the particles consists on PAH molecules. PAH molecules grow readily forming larger molecules through chemical processes and coagulation. Increase in the flame temperature causes the pyrolysis process, in which the molecules are decomposed into smaller ones. Therefore the operating conditions have influence on the number of particles emitted by the combustion process. Thus, under conditions in which high temperatures are achieved in the combustion chamber, decomposition occurs to a greater extent and hence further ultrafine particles are emitted. Under conditions of oxygen deficit and low temperature, oxidation processes are incomplete. These conditions foster the growth of ultrafine particles via condensation mechanisms, and as a result, larger particles are formed [37]. In order dissimilar to that observed for the mass of particles emitted, some studies report smaller particle number with decreasing combustion efficiency [e.g. 26].

For pine combustion, the highest number concentration was observed for the operation with low load (3.80×10^6 Ncm^{-3}·particles) during the flaming combustion phase, whilst the smallest was registered for the operation with medium fuel loads, in the heating stage ($1.15 \times 10^6 \pm 1.52 \times 10^6$ Ncm^{-3}·particles). For beech combustion, the particle number concentration was maximum during the operation with medium load ($2.91 \times 10^6 \pm 2.32 \times 10^5$ Ncm^{-3}·particles). This concentration was observed during the flaming combustion stage. The particle number concentration was minimal for the operation with high load (NS) during the flameless stage ($1.66 \times 10^6 \pm 3.39 \times 10^5$ Ncm^{-3}·particles). As regards to ignition techniques, for pine, it was verified that the particle number concentration was maximal when ignition from the bottom was performed (4.01×10^6 Ncm^{-3}·particles). This concentration was observed during the flaming combustion stage. Beech combustion showed the same trend, albeit with a slightly lower concentration ($3.74 \times 10^6 \pm 1.58 \times 10^5$ Ncm^{-3}·particles). The minimum concentrations were observed for the heating step for the top ignition test ($1.01 \times 10^6 \pm 6.31 \times 10^5$ Ncm^{-3}·particles). For the combustion of beech, the lowest concentrations were recorded during the flameless combustion step (char combustion), when the experiment with top ignition was carried out ($1.96 \times 10^6 \pm 2.78 \times 10^5$ Ncm^{-3}·particles). Johansson [38] investigated the particulate emissions from two residential combustion devices, including a stove and a pellet boiler. The PM_{10} number concentrations ranged between 1.8 and 8.7×10^7 Ncm^{-3}·particles. The highest particle number concentrations are associated with good combustion conditions, easily achieved in devices with advanced combustion technologies.

In the present work, a relationship between the temperature in the combustion chamber and the particle number was observed. The increase in the combustion temperature leads to the emission of a larger number of particles. Wardoyo [37] conducted tests on a modified combustion stove to allow the introduction of a controlled amount of air and variable rates of combustion. The author aimed to evaluate the influence of temperature on the number concentration of particles resulting from the combustion of hardwoods. Two combustion conditions were tested: "vigorous combustion" (high combustion air flow rate) and "smouldering combustion" (restricting the combustion air intake). As seen in this study, the author found that, in all measurements performed, the particle number concentration maximized for higher temperatures. The author found a linear relationship (R^2>0.93) between temperature and particle number concentration for tests conducted under "vigorous combustion" conditions. For the "smouldering combustion" conditions a linear relationship was also found, although large fluctuations in particle number concentrations have not led to a statistically significant relationship. The author found that the "vigorous combustion" conditions generate higher particle number concentrations and temperatures than those of the "smouldering" conditions.

3.6 Number emission factors

For pine combustion, the maximum global emission factor was recorded when operating with low load (1.03×10^{17} particles·kg^{-1} biomass), whereas the minimum was registered when operating with non-split logs in high load conditions ($4.08 \times 10^{16} \pm 3.48 \times 10^{15}$ particles·kg^{-1} biomass). Beech combustion showed the same trend described above. The overall maximum emission factor was observed when operating with low load ($7.80 \times 10^{16} \pm 1.08 \times 10^{16}$ particles·kg^{-1} biomass), whereas the minimum occurred under conditions of high load (S) ($2.47 \times 10^{16} \pm 6.09 \times 10^{14}$ particles·kg^{-1} biomass).

Regarding the ignition technique, it can be seen that, for both woods, the overall emission factor is higher when the ignition is carried out from the top, although in the case of beech combustion the difference between techniques is negligible. The top-down ignition technique generated emission factors between $5.26 \times 10^{16} \pm 1.15 \times$

10^{16} and $3.68 \times 10^{16} \pm 2.54 \times 10^{15}$ (particles·kg^{-1} biomass) for pine and beech, respectively. The bottom ignition technique contributed to emission factors between 5.72×10^{15} and $3.69 \times 10^{16} \pm 2.33 \times 10^{15}$ (particles·kg^{-1} biomass) for pine and beech, respectively. The top ignition leads to a progressive combustion of the fuel, resulting in more complete combustion, as described above. Thus, the increased combustion efficiency leads to number emission factors exceeding the number recorded for the traditional lighting technique, which originates incomplete combustion conditions. The various tests with various biomass loads led to higher numerical emission factors for pine than for beech.

Tissari et al. [39] studied the influence of the type of equipment and operating conditions on the particle number emission factors. The experiments involved the use of combustion appliances typically employed in Finland (conventional masonry stove and modern stove). Different biomass loads and log sizes were evaluated. The authors concluded that the number emission factor was higher for the modern combustion equipment (5.9×10^{14} particles·kg^{-1} biomass) than that obtained with the conventional stove (3.1×10^{14} particles·kg^{-1} biomass). Moreover, the authors found that higher loads produce smaller number of particles (2.0×10^{14} particles·kg^{-1} biomass) in comparison with low loads (4.0×10^{14} particles·kg^{-1} biomass). This trend was also observed in the present study, although the recorded values are higher. The variations are probably related to differences in characteristics of combustion devices. When the degree of cleavage applied was higher, i.e. when smaller wood pieces were used, the authors found an increase in the number of particles (3.1×10^{14} particles·kg^{-1} biomass) comparatively to the number of particles emitted during the combustion of larger pieces of wood (2.2×10^{14} particles·kg^{-1} biomass) [39].

Tissari et al. [26] conducted tests on a conventional masonry stove. These tests comprised "normal combustion" and "smouldering combustion" conditions. The slow combustion conditions have been created using an excess of fuel load and restricting the entry of combustion air. The authors found that in improper combustion conditions (i.e. with excess of fuel and oxygen deficit in the combustion chamber), the number of particles was about 20% lower compared to "normal" combustion conditions. The smaller number of particles occurs due to coagulation and condensation that contribute to the growth of the particles. Thus, the particle number is reduced and, moreover, size increases. The authors found that the mass emission was quite high due to the large sizes of particles emitted. Wardoyo [37] investigated the emissions of particles in a modified stove in order to allow the introduction of a controlled amount of air and variable combustion rates. The author tested "vigorous combustion" and "smouldering combustion" conditions. The particle number for the "vigorous combustion" conditions ranged from 3.3 to 5.7×10^{15} (particle·kg^{-1} biomass). The "smouldering" conditions generated a lower number of particles (from 2.8 to 44.8×10^{13} particles·kg^{-1} biomass), as observed by Tissari et al. [26]. The tests performed in this study produced fewer particles for high load conditions with non-split logs, for which the combustion air supply is insufficient for high devolatilization rates of biomass. Note that, for operation under low load, the PM mass emission factors were the highest among the tested conditions. As expected, these results were associated with unfavourable combustion conditions with reduced temperatures and low combustion rates. It should be taken into account, however, that the exact replication of conditions between successive combustion experiments is difficult, which causes variations in results.

3.7 Aerosol size distributions

When operating with reduced load, it was observed that dg is higher during the flaming phase of combustion for both pine (0.206 µm) and beech (0.254 ± 0.045 µm). For the operation with medium load, dg was higher during the flaming combustion stage, when pine was used as fuel (0.205 ± 0.008 µm), or during the start-up phase, when beech was burned (0.310 ± 0.139 µm). In the case of high load operation without splitting the pine logs, the dg was higher for the start-up phase (0.430 ± 0.069 µm). For beech combustion, the highest dg was observed during the flaming phase (0.282 ± 0.148 µm). For both woods, the highest dg were obtained for high load operation. Regardless of the load, the smallest diameters were recorded during the smouldering phase of combustion for both woods, with values between 0.110 ± 0.024 (pine) and 0.122 ± 0.002 µm (beech).

With regard to the ignition technique, it has been found that, for pine combustion, the highest dg were recorded in the heating up phase, when using the top ignition technique (0.197 ± 0.078 µm) or in the flaming phase when the bottom ignition technique (0.191 µm) was tested. For beech combustion, the highest dg were registered in the heating up phase either when using the top ignition technique (0.245 ± 0.007 µm) or bottom ignition (0.294 ± 0.038 µm).

The particle size increases during conditions of incomplete combustion. Tissari et al. [39] found that modern biomass combustion equipment emit more particles, but smaller diameters. They also observed that the use of high biomass loads causes the increase of the particle diameter. This trend was also observed when using small pieces of wood. Pettersson et al. [22] concluded that the particulate material resulting from domestic combustion processes is dominated by submicron particles in the range from 0.1 to 0.3 µm. The researchers also found differences in the size distributions for the distinct stages of the combustion cycle. In this work, these trends were also observed. Conditions of incomplete combustion cause the formation of larger aggregates of soot and condensation of organic material into the pre-existing particles during the cooling of the exhaust effluent, modifying the particle diameters [22]. The increase of particle diameters under conditions of incomplete combustion was also observed by other authors. Tissari et al. [26] described a 2.5-fold increase in the geometric mean diameter for incomplete combustion conditions when comparing with "normal" combustion conditions.

A maximum number of 9.6×10^{6} particles·cm^{-3} for the size bin of 0.28 µm was observed during the first stage of the combustion cycle with medium fuel load. Subsequently, there was a progressive decrease in the number of particles collected on the following channels. During the flaming stage, when the number of particles is at its maximum, a peak of 1.9×10^{7} particles·cm^{-3} for the size bin of 0.22 µm was registered. During the flameless combustion stage, the peak, 7.1×10^{7} particles·cm^{-3}, was recorded for the first channel (0.08 µm). At this stage, the number of particles within the smallest size ranges was experienced an increase compared with the values observed for phases 1 and 2. In the case of beech, the evolution of the number of particles is similar to that described above for pine combustion. In the first stage of the combustion cycle, a peak of 8.1 x10^6 particles·cm^{-3} in the size channel of 0.31 μm was recorded. Thereafter, there is a progressive reduction in the number of particles. During the flaming phase, a maximum of 1.3 × 10^7 particles·cm^{-3} in the size channel of 0.25 μm was achieved. For flameless combustion, the largest number, 6.5 × 10^6 particles·cm^{-3}, was recorded for the third channel (0.13 μm). At this stage, an increase in the number of particles within the smallest size bins is observed compared to the values of phases 1 and 2.

4 CONCLUSIONS

The present work had as objective to experimentally characterize and quantify the gaseous and particulate emissions during combustion in a residential woodstove with variations in fuel and operating conditions. The comparison of values from this study with literature data showed differences, depending mainly on operating procedures and combustion technologies. Thus, to implement measures and as a guideline for future regulations, emission factors from different types of combustion devices under different operation modes are needed. This work may be potentially useful because it provides some guidance on best practices for households and end-users in Southern European countries, where the combustion of wood in traditional appliances is becoming ever more common.

The analysis of the CO and THC evolution over a combustion cycle showed that the initial stage is critical from the point of view of unburnt compound emissions, due to the rapid devolatilization of biomass in a short period of time. The lowest particle emissions were generated under operation with medium fuel loads, whereas the highest were achieved for low and high loads of pine and beech, respectively. Cleavage also demonstrated to play an important role. Particulate emissions increase with the decrease of the surface area of the logs. Regarding the ignition technique, it was observed that the top-down ignition technique can reduce the PM_{10} emission factor to less than half compared with the traditional technique. The OC content was higher when operating under high and low load conditions using pine and beech as fuels, respectively. The top-down ignition technique generated substantial amounts of EC. The lowest OC/EC ratios were observed for the top-down ignition technique, whilst the highest values corresponded to the operation with non-split logs of both woods. The highest particle number emission factors and the smallest particle geometric mean diameters were reached under low fuel load operation. The use of non-split logs generated, for both woods, the highest dg, with values up to 0.39 μm.

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