Particulate and gaseous emissions from residential pellet combustion

Estela Vicente, Márcio Duarte, Teresa Nunes, Luís Tarelho, Célia Alves

Centre for Environmental and Marine Studies, Department of Environment and Planning, University of Aveiro, Portugal

E-mail: estelaavicente@ua.pt , ltarelho@ua.pt

ABSTRACT

Seven biomass fuels were tested in an automatic pellet stove (9.5 kW) in order to determine emission factors of gaseous compounds, such as carbon monoxide (CO), nitrogen oxides (NO_x) and hydrocarbons. The particulate matter (PM_{10}) emission factors for each fuel were also obtained, as well as the organic (OC) and elemental carbon (EC) content in PM_{10}. The fuel type clearly affected the gaseous and particulate emissions. The CO emission factors ranged from 91.1±19.4 (pellets type IV) to 1477±125 mg MJ^{-1} (olive pit). The NO emission factors were between 89.3±8.72 (olive pit) and 307±80.1 mg MJ^{-1} (pellets type II). The NO_2 emission factors ranged from 0.10±0.08 (pellets type IV) to 9.09±2.48 (pellets type III) mg MJ^{-1}. The variability of NO_x emissions among fuels can be explained by the variation in the nitrogen content of fuels. Regarding hydrocarbons, wood pellets had the lowest emission factor when compared with the other fuels; olive pit was the fuel with highest emission factor. The PM_{10} emission factors ranged from 24.6±2.90 to 156±21.8 mg MJ^{-1}. The lowest PM_{10} emission factor was found for wood pellets type I (fuel with low ash content) while the highest was observed during the combustion of an agricultural fuel (olive pit); the highest emissions were generally observed for the agricultural fuels. The OC content of PM_{10} ranged from 8 wt.% (pellets type II) to 29 wt% (olive pit). Variable EC mass fractions in particulate matter, ranging from 3 wt.% (olive pit) to 47 wt.% (shell of pine nuts), were also observed. The carbonaceous content of particulate matter was lower than that reported previously during the combustion in traditional woodstoves and fireplaces.

Keywords: *Residential combustion, Emission factor, Pellets, Agricultural fuels, Stove.*

1. INTRODUCTION

Biomass combustion has been encouraged in order to reduce fossil fuel consumption. Despite being considered renewable and CO_2-neutral, biomass combustion is recognized as a source of air pollution. High emissions from incomplete fuel combustion in small-scale appliances like woodstoves and fireplaces have been reported in several countries [1–3]. However, other technologies are available for domestic heating purposes with advantages from the emission point of view. Small scale pellet heating systems are installed in rising tendency. The wood pellet market has experienced a large growth in recent years as a result of the EU objective to increase the share of renewable energy [4]. The rising costs of oil on the international markets also boosted the use of renewable energies as a heating source [5]. The use of pellets as fuel in small scale appliances for heating purposes has been pointed out as suitable in order to reduce the emissions from this sector [6].

In 2006 the production of wood pellets was estimated between 6 and 7 million tons worldwide, not including Asia, Latin America and Australia. In 2010 the global wood pellet production reached 14.3 million tons, whist the consumption was close to 13.5 million tons, thus registering an increase of more than 110% compared to 2006. Between 2008 and 2010 the production of wood pellets in EU increased by 20.5%, reaching 9.2 million tons in 2010, representing 61% of the global production. In the same period, the EU wood pellet consumption increased by 43.5% to reach over 11.4 million tons in 2010, representing nearly 85% of the global wood pellet demand. In the segment of residential heating, the main drivers for pellets market expansion are often indirect support measures for the installation of pellet stoves and boilers, as well as the relative cost competitiveness of wood pellets compared to traditional fuels, such as LPG heating oil and natural gas, especially in rural areas that are not yet supplied by gas grids [7]. For example, in Spain, the Ministry of Economy has encourage the installation of boilers as part of the 2004-2012 Energy Plan, which aimed to promote the use of biomass, such as pellets, olive pit and almond shell, as an energy source. In Portugal, Spain and other southern European countries, the cork and olive oil sectors generate large amounts of residues that can be used as fuel for heating in small scale appliances [8]. In Portugal, the potential for producing pellets through the use of agricultural residues is recognized. The energy potential derived from almond residues, for example, was estimated to be 93 kton y^{-1}. Cereal straw and pruning residues are the agro-residues with more energy potential [9]. Although good combustion conditions lead to lowest particulate emissions, studies performed, in healthy C57BL/6J mice and RAW264.7 macrophage cell line, have reported highest oxidative stress, inflammatory, cytotoxic and genotoxic activities and a decreased cellular metabolic activity from particles generated under efficient combustion conditions rather than particles resulting from smoldering combustion [10, 11]. In spite of the increasing use of pellet stoves, their emissions are poorly typified. The variability in properties of biomass is great and may significantly influence the efficiency and environmental impacts associated with their use, constituting an issue of great importance and research interest.

The aim of this research is to characterize the gaseous and particulate emissions resulting from a Portuguese model of pellet stove with growing market share in the residential sector, where different solid biofuels (four types of pellets, olive pit, almond shell and shell of pine nuts) have been burnt. The work comprises extensive quantitative and qualitative data for gaseous compounds and particulate matter (PM_{10}). The organic carbon (OC) and elemental carbon (EC) content of PM_{10} was quantified.

2. EXPERIMENTS

2.1 Appliance, fuels and experimental procedures

The combustion experiments were carried out using a top-feed pellet stove with a nominal output of 9.5 kW, model Alpes, manufactured in Portugal by Solzaima.

The stove has internal pellet storage with 20 kg of capacity and the fuel is supplied by an auger screw to the burner. The primary air is supplied through holes in the bottom of the grate driven by an electric fan. The secondary air is feed above the grate through three holes by natural convection. Primary and total combustion air flows were monitored during the combustion cycle. Temperatures were measured continuously using K-type thermocouples at several locations along the system (combustion chamber, chimney and dilution tunnel). The stove can be operated at five levels of power output by automatically modifying the fuel feed rate and the fan speed. In order to cover different behaviors by users, the emission factors were determined for three levels of power output (lowest, medium and highest). The ignition of the fuel is made through an electrical resistance heater and air supply from a blower. The ignition phase was not included in the experiments.

It should be pointed out that a short cleaning period of the grate is programmed to occur. During the cleaning process, the fuel supply decreases and the air supply increases for a few minutes in order to remove the bottom ashes from the grate.

Each of the combustion experiments was performed after preheating the pellet stove. When the level of power output was changed, it took about 40 minutes to start the experiments to ensure that the combustion process had already stabilized.

The feeding rate for each fuel was estimated prior to the experiments in the three levels of power output. For this purpose pre-weighed pellets were poured into the hopper and after burning for about 2 hours the remaining fuel was re-weighed.

The stove was connected to a chimney equipped with a sampling port for flue gas sampling and characterization.

To investigate the influence of fuel quality on gaseous and particulate emissions, four types of pellets were selected. Pellets type I were commercial wood pellets made of golden wattle and pine. Pellets type II were composed of 75% of lignocellulosic residues and 25% of dust from the furniture manufacturing industry. Pellets type III were composed of 65% of lignocellulosic residues and 35% of dust from the furniture manufacturing industry. Pellets type IV were made with a mixture of 50% of waste woodchips (several woods from construction and demolition, pine wood pallets, forest biomass, paper and paperboard) and 50% of dust from the furniture manufacturing industry. In addition, emission factors for the combustion of olive pit, almond shell and shell of pine nuts were obtained. Fuel properties, including lower heating value (LHV), moisture, C, H, N, O, S and ash content are listed in Table 1. Moisture and ash content of wood pellets for non-industrial use are two of the parameters established by the European norm EN 14961-2. Some of the fuels exceeded the moisture and ash contents defined by the norm.

Table 1. Characteristics of the tested fuels

		Pellets type I	Pellets type II	Pellets type III	Pellets type IV	Olive pit	Pine nuts shell	Almond shell
Proximate analysis (wt.% , as received)	Moisture	8.4	8.8	10.9	10.7	12.9	12.9	9.5
Ultimate analysis (wt.%, dry basis)	Ash	0.73	3.2	3.8	2.0	0.66	1.3	1.4
	C	49.7	47.4	48.3	47.4	50.9	49.8	49.3
	H	6.9	6.58	6.53	6.79	6.59	6.59	6.76
	N	0.16	2.31	2.06	2.11	0.21	0.30	0.34
	S	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
	O (by difference)	42.5	40.5	39.3	41.7	41.6	42.0	42.
LHV (MJ kg^{-1})		18.3	17.5	17.4	17.8	18.5	18.7	18.4

2.2 Flue gas characterization

The combustion flue gas was extracted from the chimney by means of a heated probe and line at 180 ºC and sent to a Fourier transform infrared gas analyzer (FTIR Gasmet™, CX4000). This equipment has a multicomponent measurement capability, which enables, among others, the real-time monitoring of H_2O, CO_2, CO, NO, NO_2, SO_2, NH_3, HCl, CH_4, C_2H_4, C_2H_6, C_3H_8, C_6H_{14} and CHOH. The sample cell heating ensures even high water vapor concentrations or corrosive gases will not pose a problem. Gasmet™ CX4000 has both TÜV (17th BlmSchV) and MCERTS certificates. The analyzer is also compliant with U.S.

EPA 40 CFR Part 60 Appendix b Performance Specification 15. The advanced, easy-to-use Calcmet™ software provides outstanding analytical performance. Calcmet™ analyzes the sample spectrum using sophisticated and patent protected multicomponent algorithms. Calcmet™ is capable of simultaneous detection, identification and quantification of up to 50 different gas components. Cross-interference effects are compensated for and analysis accuracy is maintained even when analyzing complex gas mixtures where there is a possibility of spectral overlapping. Since water content of the sample gas is continuously measured with the Gasmet™, the results can be reported on either a "wet" or "dry" basis. The results can also be compensated for the correct oxygen levels. The emission factors reported in the present work were corrected to dry gases and 13% (volume) oxygen.

2.3 Particulate matter sampling and analyses

Particulate matter (PM_{10}) was collected after the dilution of the combustion exhaust gases in a tunnel under isokinetically conditions. The sampling point was located 10 m downstream the dilution tunnel entrance. The dilution system was described in previous studies [2,12,13]. The sampling train included a PM_{10} inlet head, a pump, and a control and data acquisition system, all part of a TCR TECORA (model 2.004.01) instrument. The equipment has been operated at a flow of 2.3 m^3 h^{-1} (atmospheric temperature and pressure). The particulate matter sampling was made under steady state operating conditions, which have been evaluated by continuous monitoring of flue gas composition. The particulate matter (PM_{10}) samples for gravimetric and chemical analyses were collected on 47 mm diameter quartz fiber filters pre-baked at 500 ºC for 6 h, to remove organic contaminants. The filters were kept in a desiccator to stabilize without hydration or contamination. The gravimetric quantification was performed with a microbalance (RADWAG 5/2Y/F with an accuracy of 1 µg). Filter weight was obtained from the average of six measurements, when the variations were less than 0.02%. The organic (OC) and elemental carbon (EC) content of PM_{10} was analyzed by a thermal optical transmission technique. This method allows the differentiation of various particulate carbon fractions through the volatilization and oxidation to CO_2 under controlled heating. The CO_2 produced is then quantified in a nondispersive infrared (NDIR) analyzer. The blackening of the filter is monitored using a laser beam and a photodetector, which enables separating the EC formed by pyrolysis. A more detailed description of the method can be found elsewhere [14].

3. RESULTS AND DISCUSSION

3.1 Gaseous emissions

Gaseous emissions are greatly affected by the biofuel type (Figures 1, 2, and 3). The CO emission factors ranged from 91.1±19.4 (pellets type IV) to 1477±125 mg MJ^{-1} (olive pit). Ozgen et al. [15] tested two types of pellets (low-quality cheap pellets and high quality pellets with DIN-PLUS certification) in a pellet stove (8 kW) and pellet boiler. The authors found an average CO emission factor of 88 and 350 mg MJ^{-1} for the combustion in the stove and boiler, respectively. Verma et al. [16] used a multi-fuel boiler (40 kW) to test peat pellets, apple pellets, wheat straw pellets with hydrated lime as additive and sun flower husk pellets. The authors found that, among the agro-pellets, peat combustion generated the highest CO emissions. The CO emissions for the combustion of peat pellets were 50.0, 6.1 and 2.5 times higher than those of apple, straw and sun flower husk, respectively. In the present work, olive pit was the fuel with highest CO emission factor. The latter was about 16 times higher than that observed for pellets type IV, which produced the lowest emission factor. Miranda et al. [17] studied the emissions from the combustion of pelletized residues from olive pomace and Pyrenean oak in a pellet stove (5.8 kW) . The authors tested both fuels separately and a 50/50 blend. Emissions from the combustion of pure olive pomace pellets were slightly worse, so it was concluded that blends with more than 50% of this product are not recommended [17]. Sippula et al. [18] tested pellets of bark and stem of birch, pine, spruce, alder and willow in a pellet stove (8 kW). The lowest CO emissions were recorded for commercial, pine stem, and spruce stem pellets (101-201 mg MJ^{-1}). Higher emissions were measured from stem pellets made of alder, birch, and willow (207-355 mg MJ^{-1}). The CO emission factors obtained in the present study for the agricultural fuels are in the range of values obtained for batch combustion (390-2700 mg MJ^{-1}) [19]. Lamberg et al. [20] tested pellets made of pine steam wood as primary raw material in a pellet boiler (25 kW) operated under normal conditions and observed a CO emission factor of 63±56 mg MJ^{-1}. Operational manipulations related to the boiler load, and the reduction of primary and secondary air supplies led to CO emission factors in the range from 3.55±7.06 to 335±292 mg.MJ^{-1}.

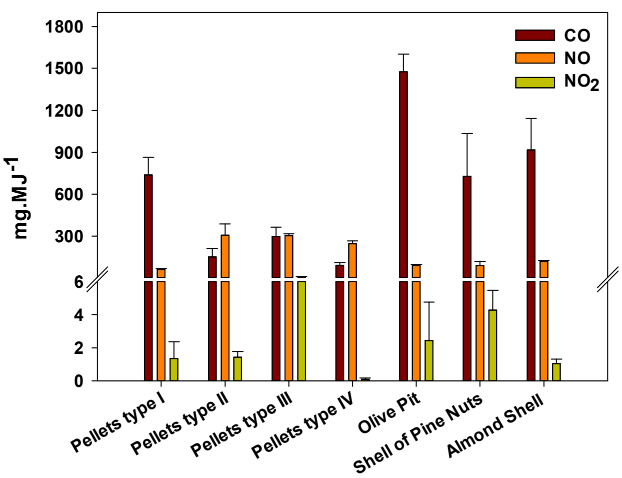

Figure 1. CO, NO and NO_2 emission factors for the combustion of different fuels (mg MJ^{-1}).

The NO emission factors observed here were between 89.3±8.72 (olive pit) and 307±80.1 mg MJ^{-1} (pellets type II). The NO_2 emission factors ranged from 0.10±0.08 (pellets type IV) to

9.09±2.48 (pellets type III) mg MJ^{-1}. The variation in NO_x emissions may be related to different nitrogen contents of fuels [16]. Fuel-NO is the main mechanism of NO formation in this type of equipment. The temperatures recorded in the combustion chamber are typically too low for the onset of the NO thermal mechanism [16,21,22]. Pellets type II, III and IV are the biofuels with highest N contents and, thus, present the highest NO_x emission factors.

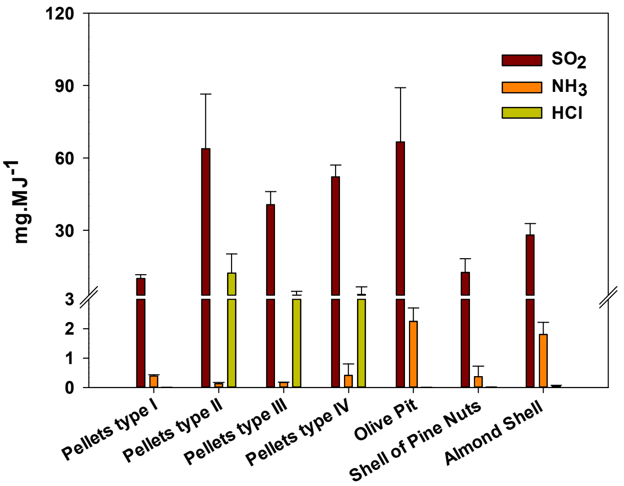

Figure 2. SO_2, NH_3 and HCl emission factors for the combustion of different fuels (mg MJ^{-1}).

González et al. [23] studied the combustion in a pellet boiler (12 kW) of residues from almond tree (pruning and almond shell and peel) and one forest biomass pellet recommended by the boiler manufacturer. The authors observed that the behavior shown by almond tree pruning and almond shell was similar to that of the forest biomass pellet. Thus, these agro-fuels can be substitutes of forest biomass pellets. In respect to gaseous emissions, this is in agreement with the results obtained in the present study.

The use of agro residues may be problematic from the point of view of operating issues and emissions (e.g. NO_x and SO_2) if the content of N, S and Cl is high. However, in the present work, all agro residues have low contents of N and S, leading to NO_x, HCl and SO_2 emissions in the ranges of those resulting from the combustion of pellets (Figures 1 and 2).

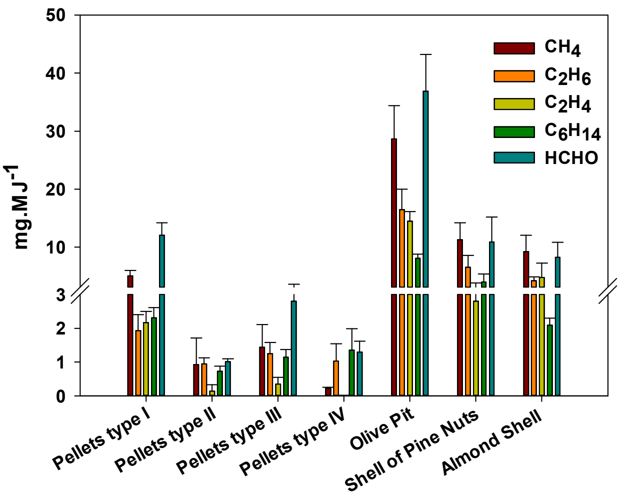

Figure 3. CH_4, C_2H_6, C_2H_4, C_6H_{14} and CHOH emission factors for the combustion of different fuels (mg MJ^{-1}).

Regarding hydrocarbons (Figure 3), wood pellets have lower emission factor (EF) than the other fuels studied; olive pit was the fuel with highest EF for hydrocarbons. This is possibly related to the fact that olive fruit has a stony pit containing 30% lipids and 20% carbohydrates.

3.2 PM_{10} emission factors

Particulate matter emission from residential wood combustion for heating purposes is one topic of great concern. The technical characteristics of residential combustion appliances play a major role in emissions. Appliances may be divided into two groups: manually fed and batch-operated combustion systems, and automatically fed systems. The fuel properties may also have a significant influence on the amount and composition of particle emissions [3,18–20,24].

PM_{10} emission factors from the combustion experiments are listed in Table 2. The emission factors ranged from 24.6±2.90 (pellets type I) to 156±21.8 mg.MJ^{-1} (olive pit). Globally, the PM_{10} emission factors observed in the present study are 8 to 10 times lower than those obtained in a traditional fireplace [12], [24]. In general, the highest EF were obtained for biomass fuels other than pellets, suggesting that the fuel particle size can affect the biomass feeding rate into the stove, which, in turn, can influence the equipment performance and its emissions.

Table 2. Fuel feed rate and PM_{10} emission factors for the biomass fuels tested.

		Fuel feed rate (kg h^{-1})		PM_{10} (mg MJ^{-1})	
Fuel	Number of experiments	average	std.	average	std.
Pellets type I	4	1.18	0.17	24.55	2.90
Pellets type II	9	1.40	0.23	79.90	12.54
Pellets type III	7	1.12	0.17	94.05	7.96
Pellets type IV	6	1.44	0.20	69.91	8.68
Olive Pit	6	0.88	0.07	156.07	21.81
Shell of Pine Nuts	8	0.96	0.18	108.28	31.37
Almond Shell	6	1.36	0.29	103.82	3.74

Studies have been performed in order to quantify and characterize the particulate emissions from pellet stoves. Figure 4 shows a comparison between the results obtained in previous studies using distinct appliances and the results obtained in the present work. Open fireplaces present the highest particulate emission factors; the low temperatures in these appliances contribute to inefficient combustion, and smoldering is the prevalent combustion phase. The continuous improvement of combustion technologies has contributed to significant reduction in emissions.

Fernandes et al. [24] found $PM_{2.5}$ emission factors of 707±255, 446±133 and 98.7±44.8 mg MJ^{-1} for wood combustion in a fireplace, woodstove and eco-labeled woodstove, respectively (Figure 4). Even automatically fed stoves and boilers may present varying emissions. Although the emissions resulting from the combustion in automatically fired appliances are significantly lower than those from log-woodstoves and fireplaces, variations can be observed. These variations may be associated either with the fuel properties or with the burning mode (full or part load operation) [3,8,20,25].

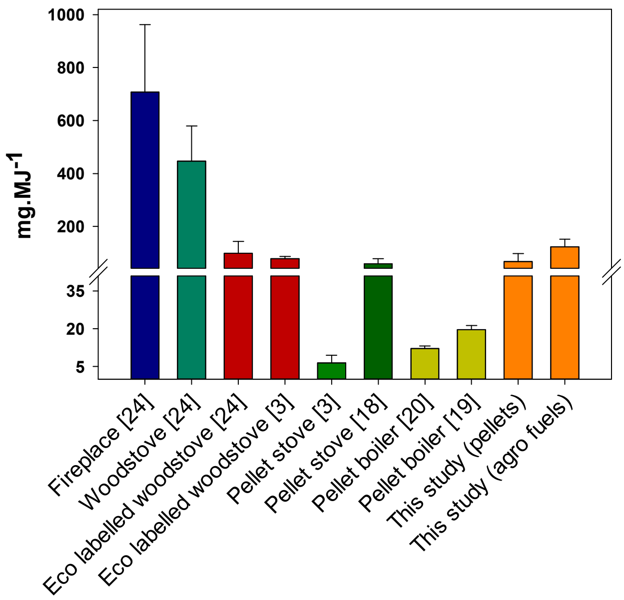

Figure 4. Particulate matter emissions from different types of small-scale combustion appliances (mg MJ^{-1}).

Garcia-Maraver et al. [8] found that gaseous and PM emissions are significantly affected by the fuel type used rather than the boiler thermal load. Ozgen et al. [15] tested two types of pellets (low quality cheap pellets and high quality pellets with DIN-PLUS certification) in a pellet stove (8 kW) and boiler (25 kW). The authors reported an average PM emission factor of 109 mg MJ^{-1} for the pellet stove and of 61 mg MJ^{-1} for the pellet boiler.

Particles resulting from residential wood combustion are composed of soot, organic matter and fine fly ash [5,26–28]. Particles from manually operated small-scale combustion appliances are, in general, dominated by organic matter, because the combustion conditions are more incomplete. Automatically fired equipments allow operating under more stable and efficient conditions. The particulate matter resulting from the combustion in these equipments is mainly composed of alkali salts. The ash content and composition of the fuel have a great influence on emissions. Thus, the variability in the PM_{10} emission factors observed in the present study for the different fuels may be explained by the different ash contents. Pellets type I showed the lowest emission factor and the lowest ash content (0.73 wt.%), while pellets type III exhibited the highest emission factor and ash content (3.8 wt.%). Sippula et al. [18] found that the fuel ash content correlated linearly with the PM_1 emission.

Figure 5 shows the PM_{10} carbonaceous content for the distinct combustion experiments. Total carbon represented 17.0 (almond shell) to 62.6 wt.% (shell of pine nuts) of the PM_{10} mass. The OC content of PM_{10} ranged between 8.2 (pellets type II) to 28.9 wt.% (olive pit), while EC represented 3.1 (olive pit) to 46.6 wt.% (shell of pine nuts) of the PM_{10} mass. The carbonaceous content of particulate matter was lower than that obtained during the combustion in traditional woodstoves and fireplaces [12].

Leskinen et al. [29] studied the physico-chemical properties of particles emitted under three different combustion conditions, namely efficient, intermediate and smoldering combustion. In order to produce the three different combustion conditions the authors used a biomass combustion reactor with a moving grate. Considerable differences between combustion conditions were observed. PM_1 from the efficient combustion conditions consisted almost entirely of ash-related material. Under the intermediate conditions the particulate matter was composed mainly of OC and EC in nearly equivalent portions. For smoldering conditions, the particles emitted contained more EC than OC.

Improved combustion efficiency with higher combustion temperatures and flaming combustion contribute to higher EC emissions than those resulting from the combustion in equipments with lower efficiency like fireplaces and traditional woodstoves [24]. Gonçalves et al. [2] tested several fuels in a "chimney type" logwoodstove (6 kW) and found that total carbon represented 43.9-63.2 wt.% of the $PM_{2.5}$ mass. Schmidl et al. [30] observed higher total carbon content in the particulate matter emitted from traditional manually fired appliances, with EC contributing to about 30% and e OC to 30-40% of the PM mass.

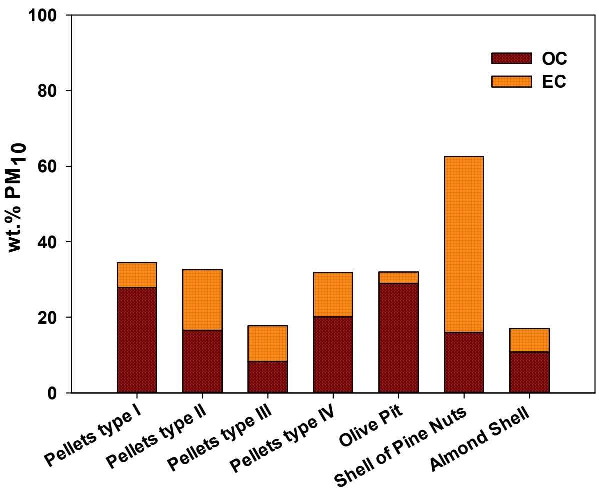

Figure 5. Carbonaceous components in wood smoke (wt.% PM_{10}).

Depending on solid biofuels, different mass fractions of OC and EC were observed in the emitted particles in this study (Figure 5). The combustion of shell of pine nuts generated higher amounts of EC in the particulate matter than OC, while the opposite was observed for olive pit. The latter was the fuel with highest hydrocarbon emission factors, as described above. Hydrocarbons condense onto existing particles, likely contributing to the high OC content.

Primary soot particles are formed mainly in the flame from hydrocarbons combustion. Depending on the temperature and oxidative conditions, primary particles may be burned or remain in the particulate phase. As a consequence of the insufficient mixing of combustion gases and air, the flame zone always contains fuel-rich areas providing the conditions for the formation of soot particles [26,28,31]. This mechanism may be the explanation for the relatively high EC content in the particulate matter from the combustion of pine nuts shell. The varying carbon fractions are thus related to temperature, mixing and oxygen supply.

4. CONCLUSIONS

This paper constitutes an attempt of quantifying particle and gaseous emissions from combustion of different solid biofuels in a pellet stove. It was found that the fuel type and its properties can significantly influence the gaseous and particulate emission factors from residential combustion equipments. In addition, through the organic and elemental carbon mass fractions, it was possible to verify that the chemical composition of particles can also vary noticeably.

Combustion in modern appliances can reduce PM emissions significantly when compared to old types of wood-fired equipments.

The CO emission factors were lower for pellets than for agro residues. However, the considered higher quality commercial pellets showed the highest CO emission factor among all pellet types. The NO emission factors were found to be lower when testing an agricultural fuel (olive pit) and the NO_2 when burning pellets made of waste woodchips and dust from the furniture manufacturing industry (pellets type IV). The use of agricultural fuels may be problematic from the point of view of emissions if the content of N, S and Cl is high. However, in the present work it was observed that all tested agro-residues have low contents of N and S keeping low the emissions of NO_x and SO_2. Regarding hydrocarbons, wood pellets had the lowest emission factor when compared with the other fuels.

The PM_{10} emission factors were lower for the combustion of pellets with lower ash content (type I) and higher when the agro-fuels were burned. The fuel ash content was found to be a parameter with great influence on particulate emissions. Combustion of the different fuels generated differences in the OC and EC contents in the particles emitted. Thus, the fuel properties can have a significant influence on the combustion efficiency even when using automatically fired equipments.

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