



## Particulate and gaseous emissions from the combustion of different biofuels in a pellet stove



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### HIGHLIGHTS

- The highest emission factors were obtained for agro-fuels.
- Organic carbon contributed no more than 30% of the PM<sub>10</sub> mass.
- Mannosan and galactosan were not detected in almost all samples.
- Treated wood in pellets generated high contents of Pb, Zn and As in PM<sub>10</sub>.

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### ABSTRACT

Seven fuels (four types of wood pellets and three agro-fuels) were tested in an automatic pellet stove (9.5 kW<sub>th</sub>) in order to determine emission factors (EFs) of gaseous compounds, such as carbon monoxide (CO), methane (CH<sub>4</sub>), formaldehyde (HCHO), and total organic carbon (TOC). Particulate matter (PM<sub>10</sub>) EFs and the corresponding chemical compositions for each fuel were also obtained. Samples were analysed for organic carbon (OC) and elemental carbon (EC), anhydrosugars and 57 chemical elements. The fuel type clearly affected the gaseous and particulate emissions. The CO EFs ranged from 90.9 ± 19.3 (pellets type IV) to 1480 ± 125 mg MJ<sup>-1</sup> (olive pit). Wood pellets presented the lowest TOC emission factor among all fuels. HCHO and CH<sub>4</sub> EFs ranged from 1.01 ± 0.11 to 36.9 ± 6.3 mg MJ<sup>-1</sup> and from 0.23 ± 0.03 to 28.7 ± 5.7 mg MJ<sup>-1</sup>, respectively. Olive pit was the fuel with highest emissions of these volatile organic compounds. The PM<sub>10</sub> EFs ranged from 26.6 ± 3.14 to 169 ± 23.6 mg MJ<sup>-1</sup>. The lowest PM<sub>10</sub> emission factor was found for wood pellets type I (fuel with low ash content), whilst the highest was observed during the combustion of an agricultural fuel (olive pit). The OC content of PM<sub>10</sub> ranged from 8 wt.% (pellets type III) to 29 wt.% (olive pit). Variable EC particle mass fractions, 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 of several wood fuels in traditional woodstoves and fireplaces. Levoglucosan was the most abundant anhydrosugar, comprising 0.02–3.03 wt.% of the particle mass. Mannosan and galactosan were not detected in almost all samples. Elements represented 11–32 wt.% of the PM<sub>10</sub> mass emitted, showing great variability depending on the type of biofuel used.

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### 1. Introduction

Biomass combustion has been encouraged aiming at reducing fossil fuel consumption. However, high emissions from incomplete

fuel combustion in small-scale appliances like woodstoves and fireplaces have been reported in several countries (Fine et al., 2004; Gonçalves et al., 2010; Schmidl et al., 2008). 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

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to increase the share of renewable energy (Goh et al., 2012). 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 (Pettersson et al., 2010). 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 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 (Cocchi et al., 2011). For example, in Spain, the Ministry of Economy has encouraged 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 (Garcia-Maraver et al., 2014). In Portugal, the potential for producing pellets through the use of agricultural residues is recognised. 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 (Monteiro et al., 2012). Agricultural fuels present different physical and chemical characteristics compared with woody fuels (Verma et al., 2011; Obernberger and Thek, 2004). For that reason the combustion of these fuels in small scale appliances can be a challenge (Carvalho et al., 2008). Pellet appliances are sensitive to variations of the ash-forming elements in the fuel due to slag formation on the burner, which is problematic with respect to combustion efficiency (Öhman et al., 2004), due to critical inorganic elements, such as alkaline metals, that give rise to increased particulate emissions (Carvalho et al., 2013). Ash accumulation and slag formation can even lead to shut down of the appliance (Carvalho et al., 2008). The chemical composition of the fuels has also influence on the ash melting behaviour, deposit formation and corrosion (Obernberger and Thek, 2004).

Although good combustion conditions lead to lowest particulate emissions, several studies have reported highest oxidative stress, inflammatory, cytotoxic and genotoxic activities and decreased cellular metabolic activity from particles generated under efficient combustion conditions rather than particles resulting from smouldering combustion (Happo et al., 2013; Uski et al., 2014). Besides the health effects, biomass combustion particles are efficient cloud condensation nuclei and can influence the formation of precipitation (Pöschl, 2005; Rose et al., 2010). 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 research interest.

The aim of this research is to characterise the 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 ( $PM_{10}$ ). Particulate samples were analysed for organic (OC) and elemental carbon (EC), anhydrosugars and 57 elements. A comparative analysis of particulate phase emissions of parent polycyclic aromatic hydrocarbons (PAHs) and their derivatives (alkyl-PAHs, oxygenated-PAHs, azaarenes and nitrated-PAHs) from the combustion of these biofuels in the pellet stove with those from

a manually fired appliance can be found elsewhere (Vicente et al., in press).

## 2. Experimental work

### 2.1. Combustion equipment, fuels and experimental procedure

The combustion experiments were carried out using a top-feed pellet stove with a nominal output of 9.5 kW, manufactured in Portugal by Solzaima, model Alpes (Fig. 1). The stove has an internal pellet storage tank with 20 kg capacity and the fuel is supplied by an auger screw to the top feed burner. The primary air is supplied through holes in the bottom of the grate, and secondary air is feed above the grate through three holes. Both primary and secondary air are driven by an electric fan located downstream the combustion chamber. The primary air flow rate was monitored continuously during the combustion cycle using a mass flow meter. The temperature was measured continuously using K-type thermocouples located at several points along the combustion and exhaust 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 exhaust gas fan speed. In order to cover different behaviours by users, the emission factors (EFs) of distinct gaseous components and particulate matter ( $PM_{10}$ ) were determined for three levels of power output (lowest, medium and highest). Since the differences in emissions between different levels of power output for the same fuel were not statistically significant, average EFs were obtained for each compound. The ignition of the fuel is made through an electrical resistance located on the grate of the stove. The ignition phase was not included in the results.

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 combustion air supply increases for a few minutes in order to remove the bottom ashes from the grate; thus, the lighter ash fractions are then carried out with flue gas.

Each of the combustion experiments was performed after a preheating period of the pellet stove. When the level of power output was changed, it took about 40 min to start the experiments to ensure that the combustion process had already attained a new steady operation condition.

The feeding rate for each fuel was evaluated by prior calibration of the screw feeding system for the three levels of power output. Also, in all experiments, pre-weighed fuel was poured into the fuel hopper and after combustion the remaining fuel was re-weighed, in order to verify the fuel consumption rate. The feed rate showed great variations among fuels (Table 2). This can be related to the differences in the physico-mechanical properties of the fuels, such as the diameter and length, the bulk density, the fine content and the mechanical durability (Carvalho et al., 2013; Verma et al., 2011).

To investigate the influence of fuel quality on emissions, four types of pellets were selected. These biofuels were selected based on their commercial availability in order to represent the wide range of pellets on the Portuguese market. Pellets type I were commercial wood pellets made of golden wattle, cedar 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, EFs for the combustion of olive pit, almond shell and shell of pine nuts were obtained. This characterisation was carried out at

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