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Influence of operating conditions on chemical composition of particulate matter emissions from residential combustion

E.D. Vicente [a], M.A. Duarte [a], A.I. Calvo [b], T.F. Nunes [a], L.A.C. Tarelho [a], D. Custódio [a], C. Colombi [c], V. Gianelle [c], A. Sanchez de la Campa [d], C.A. Alves [a,*]

[a] Centre for Environmental and Marine Studies, Department of Environment, University of Aveiro, 3810-193 Aveiro, Portugal
[b] Department of Physics, IMARENAB, University of León, 24071 León, Spain
[c] Regional Centre for Air Quality Monitoring, ARPA Lombardia, 20129 Milan, Italy
[d] Universiy of Huelva, Department of Geology, Campus Universitario de la Rábida, La Rábida, 21819, Huelva, Spain

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ABSTRACT

Wood combustion experiments were carried out in a Portuguese woodstove to determine the effects of biofuel type, ignition technique, biomass load and cleavage, as well as secondary air supply, on the chemical composition of particles (PM_{10}). Two typical wood fuels in the Iberian Peninsula were tested: pine (*Pinus pinaster*), a softwood, and beech (*Fagus sylvatica*), a hardwood. PM_{10} samples were analysed for organic and elemental carbon (OC and EC), levoglucosan and 56 elements. Total carbon (TC) represented 54–73 wt.% of the particulate mass emitted during the combustion process, regardless of wood species burned or operating condition tested. The carbonaceous component of PM_{10} was dominated by OC. The OC content of PM_{10} was higher when higher loads were fed into the combustion chamber, for both fuels. EC represented from 8 to 35 wt.% of the particulate mass. OC/EC ranged from 1.1 to 6.1 (avg. 3.0 ± 1.8) for pine combustion and from 1.1 to 3.4 (avg. 2.0 ± 0.8) for beech combustion. The lowest OC/EC ratios for both woods were observed for ignition from the top. Levoglucosan was found in all samples, representing from 3.7 to 7.5 wt.% and from 4.2 to 8.9 wt.% of PM_{10} emitted from the combustion of pine and beech, respectively. The use of low loads of fuel generated high amounts of levoglucosan either for pine or beech. Altogether, trace elements obtained by ICP-MS and ICP-AES comprised from 0.46 wt.% to 1.41 wt.% and from 0.87 wt.% to 2.36 wt.% of the PM_{10} mass for pine and beech combustion, respectively. Among elements, K, Ca, Na, Mg, Fe and Al contributed to more than 75% of the total ICP-MS mass. Potassium was the major element in almost all PM_{10} samples.

1. Introduction

Identification and quantification of sources using receptor modelling as tool in many regions across Europe pointed out biomass burning as a major source contributing to the atmospheric particulate matter (PM) levels during winter time (Bernardoni et al., 2011; Viana et al., 2013; Waked et al., 2014). Source apportionment of $PM_{2.5}$ in a coastal/rural area in Portugal showed that in winter, on average, 64% of the organic carbon originates from biomass burning. This source contributes at least to 21% of the $PM_{2.5}$ mass (Gelencsér et al., 2007). In Portugal, it was estimated that, combustion of fuelwood, is a common practice for about 34% of the population. On average, 83% of the wood consumed is for household heating, while the remaining 17% is used for cooking (Gonçalves et al., 2012).

Epidemiological studies show a positive correlation between the level of ambient atmospheric particulate contamination (PM_{10} and $PM_{2.5}$) and the increase of morbidity and mortality (e.g. Arbex et al., 2004; Halonen et al., 2008; McCracken et al., 2012). Based on daily counts of cardiovascular mortality, Ostro et al. (2008) found evidences that several $PM_{2.5}$ constituents like EC, OC, nitrates, sulphates, potassium, copper and iron may represent important contributors to cardiovascular mortality. The aerodynamic properties of particles are also of great importance since they regulate the deposition in the human respiratory tract. Ultrafine particles (particle diameter <100 nm) are particularly harmful to human health, since they have a sufficiently small size to penetrate the membranes of the respiratory tract and enter the bloodstream or be transported by the olfactory nerves to the brain (Pöschl, 2005). Thus, health effects caused by particulate matter are dependent on its physical and chemical properties (Bølling et al., 2009), which have a clear relation with combustion appliances, fuels and combustion conditions (Happo et al., 2013; Kaivosoja et al., 2013; Lamberg et al., 2011; Uski et al., 2014; Vu et al., 2012). Aerosols generated by biomass burning, under poor combustion conditions, consist mainly of carbonaceous compounds, mostly OC and smaller amounts of EC (Reid et al., 2005; Tissari et al., 2008), while during efficient combustion conditions, particles are mainly formed by ash related material (Leskinen et al., 2014; Torvela et al., 2014).

* Corresponding author.
E-mail address: celia.alves@ua.pt (C.A. Alves).

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Besides the effects on human health, particles generated by biomass combustion may also have other significant repercussions. EC contributes to positive radiative forcing through absorption. If internally mixed, it is estimated to be the second most important promoter of global warming through direct forcing, after CO_2 (Jacobson, 2001). The positive radiative forcing of EC can be increased when it is internally mixed with species with the property of scattering light (OC and inorganic salts), forming what is called the “core-shell” arrangement (Jacobson, 2001; Moffet and Prather, 2009). Nevertheless, any increase in positive radiative forcing for aged EC is also offset by increased hygroscopicity and thus reduced atmospheric lifetime due to wet deposition in humid environments (Moffet and Prather, 2009; Stier et al., 2006). A significant fraction of OC is formed by water-soluble compounds. This hydrophilic fraction can change the atmospheric radiative balance and influence the hydrological cycle since it participates in aerosol–cloud interactions (Calvo et al., 2013).

Emissions from batch mode operated appliances vary significantly during the different stages of wood combustion (e.g. Calvo et al., 2011, 2014; Tarelho et al., 2011). Several studies pointed out the importance of the type of combustion technology, fuel quality (e.g. Alves et al., 2011; Fernandes et al., 2011; Gonçalves et al., 2010; Tissari et al., 2009) and operating practises (e.g. Leskinen et al., 2014; Tissari et al., 2008) either on particulate emissions or on their chemical composition. However, most studies report the chemical characteristics of smoke particles from different equipment in which distinct biofuels have been burned by replicating the combustion conditions.

This research aims to characterise the particulate matter (PM_{10}) emissions resulting from the combustion in a typical Portuguese woodstove with variations in fuel (pine and beech), and operating conditions. The variables evaluated were the ignition technique (upside down and bottom up lighting), secondary air supply, fuel load and, in the case of high load, split (S) and non-split (NS) logs. Samples were analysed for organic and elemental carbon (OC/EC), levoglucosan and 56 elements. The information provided in the present study complements that given by Vicente et al. (2015), in which combustion temperatures, burn rates, emissions factors (g kg^{-1} of wood burned, dry basis) of both PM_{10} and flue gases, as well as particle number size distributions, were explored.

2. Methodology

A cast iron woodstove (Solzaima, model Sahara), operated manually in batch mode and with manual control of combustion air (primary air under bed feed) was selected for the combustion experiments. The combustion chamber has a height of 0.44 m, a width of 0.59 m and a depth of 0.36 m. The stove allows air heating whether by radiation or by natural and forced convection. The combustion air enters the firebox through a regulating device located below the grate of the stove. The flow rate of primary air was monitored by an in-line mass flow meter (Kurz Model: 500-40 0.0 P-2). Natural draft is the driving force that introduces the required combustion air into the combustion zone and also moves the flue gas up and out of the chimney. As regards the stove loading, each wood batch was placed on the fixed grate at the bottom of the combustion chamber. The grate was connected to a weight sensor (DSEUROPE Model 535QD-A5) in order to measure continuously the bed mass lost during the combustion experiments. A detailed description of the combustion facility and appliance can be found elsewhere (Calvo et al., 2011, 2014; Fernandes et al., 2011).

This work is based on experiments carried out with two types of woody biomass, namely pine (*Pinus pinaster*) and beech (*Fagus sylvatica*), two common species in the Iberian Peninsula. The elemental composition, ash and moisture content of both fuels are presented in Table 1.

In order to assess which operating practices are more advantageous from the point of view of emissions, several variables were evaluated: (i) fuel load and, in the case of high load, (ii) degree of cleavage of the logs, (iii) secondary air supply, and (iv) ignition technique (upside down and bottom up lighting).

Table 1
Elemental composition (dry basis), ash and moisture content of biofuels (wt.%).

(wt.%, as received)	*Pinus pinaster*	*Fagus sylvatica*
Moisture	9.90	9.60
(wt.%, dry basis)		
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 wt.%. **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 depending on the load condition under analysis: low load (1.0–1.2 kg), medium load (1.8–2.8 kg) and high load (3.7–4.1 kg). The medium load can be taken as the “reference” condition, as it is practiced in most homes. In the case of high load, the effect of cleavage on emissions was evaluated using split (S) and non-split (NS) logs. Tests with secondary air supply were performed with batches of pine wood ranging from 2.2 to 2.8 kg, i.e. with medium loads. Tests with secondary air supply, different loads and split versus non-split wood were initiated in “hot start” conditions following the addition of wood logs on glowing embers of the same wood, when the temperatures in the combustion chamber were around 100 °C (±20 °C).

The bottom up ignition was initiated by putting the fuel load on the top of two pine-cones previously kindled. The top-down ignition technique 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 that ranged between 1.8 and 2.2 kg of wood for both fuels (medium load) and were initiated in “cold start” conditions. The total number of runs to test each technique was between three and five. In order to replicate the Portuguese householder's practices, wood logs of 30–40 cm in length, whether for split or non-split logs, were used. The diameter of the NS logs, cylindrically shaped, ranged from 7 to 15 cm at the largest cross sectional dimension. These cylindrical blocks were cut longitudinally into two split logs with half the thickness. The duration of each combustion cycle depended on the operating condition and fuel, ranging from 30 to 95 min. At the end of the combustion experiments, only a small amount of burning char remained on the grate (less than 10% of the initial mass of the batch of fuel), temperatures in the firebox were between 100 °C and 200 °C and CO_2 concentrations in the flue gases reached 4%.

The flue gas was diluted before particle sampling. A dilution tunnel for particle measurement is widely used (e.g. Fernandes et al., 2011; McDonald et al., 2000; Pettersson et al., 2011; Tissari et al., 2008) to simulate the rapid cooling and mixing that occurs when the exhaust gases are released into the atmosphere. The dilution tunnel (0.20 m internal diameter and 11 m length) was installed downstream of the chimney. The input of the dilution tunnel is wider than the output of the chimney in order to allow, at the same time, flue gas dilution and effective mixing. The dilution air is ambient air whose inlet takes place at the exit of the chimney. The dilution ratios applied to the flue gases from the woodstove were around 25:1. The volumetric gas flow rate was obtained by multiplying the flue gas velocity by the cross section of the dilution tunnel. The mean gas velocity (6.3 ± 0.1 m s^{-1}) was estimated from the differential pressure monitored by a Pitot tube and respective pressure sensor (Testo AG 808) and a K-type thermocouple. The dilution factor was estimated by dividing the gas flow in the dilution tunnel by the flow of exhaust gases. Also using the S-type Pitot tube with pressure sensor and a K-type thermocouple, the stack