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Residential wood combustion in two domestic devices: Relationship of different parameters throughout the combustion cycle

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HIGHLIGHTS

- The evolution of $PM_{2.5}$ constituents throughout the combustion cycle was studied.
- A significant correlation between NH_4^+ and levoglucosan was found.
- High combustion efficiency leads to unusual elevated K^+/levoglucosan ratios.
- OC/EC and K^+/levoglucosan ratios present a significant potential relationship.

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ABSTRACT

Logs of three common Southern and mid-European woods (*Quercus pyrenaica*, *Populus nigra* and *Fagus sylvatica*) were burned in two different combustion appliances, a fireplace and a stove. The flue gas composition was monitored continuously in the exhaust ducts of both burning appliances for total hydrocarbons (THC) and carbon oxides (CO_2 and CO). Particulate matter with aerodynamic diameter below 2.5 μm ($PM_{2.5}$) was sampled in a dilution tunnel under isokinetic conditions and chemically characterised for water soluble-inorganic ions, organic and elemental carbon and levoglucosan, mannosan, and galactosan. The evolution of the emission factors of these components throughout the combustion cycle was studied. The fireplace was the combustion appliance with the highest CO, CO_2, THC and $PM_{2.5}$ emission factors. The carbonaceous matter represented 72–84% of the particulate mass emitted, regardless of species burned and combustion devices. OC/EC ratios were higher for the fireplace than for the stove, and showed a significant potential relation with K^+/levoglucosan. The dominant water soluble inorganic ions in smoke particles were K^+, PO_4^{3-}, SO_4^{2-}, and Na^+. Anhydrosugar emissions were strongly enhanced in the start-up phase, when lower temperatures are registered, and decreased progressively until the glowing combustion phase. However, K^+ emission seems to be higher in flaming-dominated combustion at higher temperature. Statistical analyses, including the Kruskal–Wallis test, principal component analysis and Pearson correlation between emission factors, were carried out. A significant correlation between NH_4^+ and levoglucosan was found for both appliances.

1. Introduction

Residential wood combustion (RWC) is of increasing concern as it is identified as a major source of atmospheric pollution, mainly in winter, in rural (Puxbaum et al., 2007) and urban areas (Reche et al., 2012). Although its use is recommended given that enables to reduce dependency on fossil fuels, it is responsible for large impacts on air quality, as well as significant potential effects on public

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health and global climate change (Bølling et al., 2009; Vu et al., 2012). The evaluation/quantification of these emissions is not an easy task, due to the fact that emissions from wood combustion are influenced by several factors, such as the stove design, operating conditions, combustion conditions and the species of wood and their characteristics (McDonald et al., 2000). Several authors have reported particle mass emission factors (EF) of aerosols from different types of household combustion devices. A wide range or variability has been found for particulate matter (PM) emissions. EF values from uncontrolled combustion devices are approximately 10–50 times higher than those from more controlled devices (Gonçalves et al., 2010; Johansson et al., 2008, 2003). This variability has been also observed for levoglucosan ($C_6H_{10}O_5$, formed from the pyrolysis of carbohydrates, such as cellulose), commonly used as molecular marker for biomass burning. Thus, Hedberg et al. (2006) argued that the levoglucosan fraction may be highly dependent on combustion conditions and it could be risky to use solely levoglucosan as a tracer for quantitative estimation of RWC.

Not only particles, but also gas emissions vary largely (Johansson et al., 2004). In terms of indoor air quality, CO concentration is a relevant parameter due to the number of people that die accidentally every year from poisoning. CO emissions are a by-product of incomplete combustion because of malfunctioning or inappropriate use of RWC devices.

Emission factors from RWC are of great interest, especially for source apportionment and modelling studies and, at the same time, to implement effective strategies and air quality regulations (Krecl, 2008). Several studies have provided mean EF of the total combustion cycle for different combustion appliances and for different wood species. However, the number of papers focused on the evolution of the EF throughout the combustion cycle is, as far as we know, inexistent.

The present study aims to characterise gaseous and aerosol emissions for the combustion of three common Southern and mid-European woods (*Quercus pyrenaica* –oak–, *Populus nigra* –poplar– and *Fagus sylvatica* –beech) in two different combustion devices: a stove and a fireplace. A time-resolved study of the evolution of EF is presented aiming to help better understand how these emissions evolve during the combustion process and how the different parameters are related to each other, as well as to provide information for implementation of mitigation measures.

2. Material and methods

2.1. Fuel characteristics

Wood from three common Southern and mid-European woods (*Q. pyrenaica*, *P. nigra* and *F. sylvatica*) was used as fuel. The wood was cut into logs of 0.3–0.4 m in length with a total biomass burned during each cycle of around 1.7–2.0 kg (0.3–0.7 kg/wood log). The main characteristics of the solid fuels are presented in Table 1.

2.2. Experimental infrastructure and combustion experiments

Wood combustion experiments were carried out using two different combustion appliances: i) a stove (Solzaima, model Sahara), operated manually in batch mode with handheld control of combustion air, and ii) a traditional Portuguese brick open fireplace operated manually in batch mode and with no control of combustion air. Both devices are equipped with a vertical exhaust duct (chimney) with 0.20 m internal diameter and 3.30 m height. Several parameters are continuously monitored during the combustion experiments: the weight of the fuel in the burning fixed bed at the grate, the temperature at several points of the experimental infrastructure, the air flow rate entering in the combustion chamber (for the stove) or at the exit of the chimney (for the fireplace). A complete description of the experimental infrastructure, the control of operating variables and the methodology carried out during the combustion experiments can be found in Calvo et al. (2014).

The combustion flue gas composition was monitored continuously at the exit of the chimney (at 2.8 m above the exit of the combustion chamber), namely for total volatile hydrocarbons (THC), O_2, CO_2 and CO. $PM_{2.5}$ in the combustion flue gas was continuously collected in the dilution tunnel located downstream of the chimney (A, in Fig. 1). This dilution tunnel consists of a tube of circular section with 11 m length and 0.20 m internal diameter. Particulate matter sampling points were located at ~10 m downstream the dilution tunnel entrance.

The combustion of a batch of fuel lasted between 50 and 90 min. The number of filters sampled during each experiment was variable, depending on the evolution of the combustion cycle. The number ranged from 6 to 7 filters per combustion test for the stove and between 2 and 4 for the fireplace. These differences were mainly due to the higher dilution ratio applied to the flue gases from the fireplace. On the one hand, to avoid clogging, but, on the other hand to collect enough material for later analysis, filters were sequentially changed throughout the combustion cycle according to the pressure drop registered by the particulate matter sampler.

2.3. Measuring equipments

2.3.1. Total hydrocarbons and carbon oxides

Total hydrocarbons and carbon oxides (CO_2 and CO) were measured using automatic analysers with flame ionisation (Dyna-FID, model SE-310) and non-dispersive infrared (Environnement, MIR 9000) detectors, respectively. Each gas analyser was calibrated with appropriate gas on zero and span points. The hydrocarbon concentrations were determined in methane-equivalents.

2.3.2. $PM_{2.5}$ fraction

Particles ($PM_{2.5}$) were collected under isokinetic conditions in the dilution tunnel onto quartz filters (47 mm diameter) using a

Table 1
Characteristics of the biofuels used in the study.

			Q. pyrenaica	*P. nigra*	*F. sylvatica*
Proximate analysis (% wt, as received)	Moisture	Stove	9.7	6.6	7.9
		Fireplace	7.5	5.6	11.0
Ultimate analysis (% wt, dry basis)	Ash		2.94	0.53	0.42
	C		47.22	48.86	47.97
	H		5.94	6.26	6.26
	N		0.20	0.07	0.04
	S		bdl	bdl	bdl
	O (by difference)		43.70	44.28	45.31

bdl-below detection limit.