

Analysis of the chemical composition of ultrafine particles from two domestic solid biomass fired room heaters under simulated real-world use



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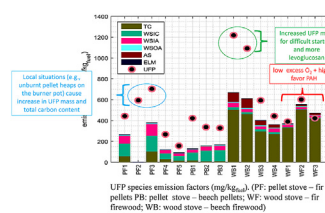
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HIGHLIGHTS

- Ultrafine particle ($d_p < 100$ nm) chemical constituent emission factors are provided.
- Automatic and manually fed room heaters with two wood types are investigated.
- Tests conducted with appliances working under real-world operating conditions.
- Increased emissions (UFP, NO_x, CO) for increased fuel ash content in the wood stove.
- PAH fingerprint shifts towards more carcinogenic PAH in wood stove UFP.

GRAPHICAL ABSTRACT



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ABSTRACT

Two common types of wood (beech and fir) were burned in commercial pellet (11.1 kW) and wood (8.2 kW) stoves following a combustion cycle simulating the behavior of a real-world user. Ultrafine particulate matter (UFP, $d_p < 100$ nm) was sampled with three parallel multistage impactors and analyzed for metals, main water soluble ions, anhydrosugars, total carbon, and PAH content. The measurement of the number concentration and size distribution was also performed by a fourth multistage impactor. UFP mass emission factors averaged to 424 mg/kg_{fuel} for all the tested stove and wood type (fir, beech) combinations except for beech log burning in the wood stove (838 mg/kg_{fuel}). Compositional differences were observed for pellets and wood UFP samples, where high TC levels characterize the wood log combustion and potassium salts are dominant in every pellet sample. Crucial aspects determining the UFP composition in the wood stove experiments are critical situations in terms of available oxygen (a lack or an excess of combustion air) and high temperatures. Whereas for the automatically controlled pellets stove local situations (e.g., hindered air-fuel mixing due to heaps of pellets on the burner pot) determine

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Wood pellet
Wood

the emission levels and composition. Wood samples contain more potentially carcinogenic PAHs with respect to pellets samples. Some diagnostic ratios related to PAH isomers and anhydrosugars compiled from experimental UFP data in the present study and compared to literature values proposed for the emission source discrimination for atmospheric aerosol, extend the evaluation usually limited to higher particle size fractions also to UFP.

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1. Introduction

Currently residential wood combustion is increasing in Europe because of rising fossil fuel prices but also due to climate change mitigation policies. However, especially in small-scale applications, residential wood combustion may cause high emissions of particulate matter (PM) (Denier van der Gon et al., 2015).

Emissions from biomass burning are found to be mostly in the fine particle size fraction. Only minor fractions of the total PM mass correspond to diameters larger than 10 μm (Boman et al., 2004). The study of the size dependent response of particle emissions to variations in combustion conditions shows that UFP contribution to the measured mass relatively increases under good operational practice with respect to the operation by restricting the combustion air supply and slightly overloading the firebox with fuel (Leskinen et al., 2014; Lamberg et al., 2011; Tissari et al., 2008). The particles are reported to contain sulfate, nitrate, chloride, sodium, potassium, calcium, magnesium, ammonium, zinc, elemental carbon, and particulate organic matter (Saarnio, 2013). Inorganic particles, mainly alkali salts, are formed from volatilized fuel ash constituents, which condense either when the temperature decreases or when chemical reactions form lower vapor pressure species (Sippula, 2010). Incomplete combustion of organic compounds formed from the decomposition due to heating of the biomass material and the consequent gas-to-particle conversion (nucleation or condensation on other existing particles) under low-temperatures is the source of organic content of the particles (Obaidullah et al., 2012). The origin of soot particles are due to reactions of tar in the early fuel-rich regions in the flame and the low temperatures in the combustion zone is a possible reason for lacking soot oxidation in the fuel-lean region of the combustion chamber with consequent release with the flue gas (Wiinikka et al., 2006). Specifically for UFP fraction, Tissari et al. (2008) found in particle samples collected from a conventional masonry heater, UFP that were composed mainly of K, S and Zn, and, to a lesser extent, of C, Ca, Fe, Mg, Cl, P and Na, while large agglomerates contained mainly carbon (soot particles). Fernandes and Costa (2012) observed for a pine pellet-fired boiler (22 kW) that UFP were composed mainly of O, K, Cl, Na and S, regardless of the boiler operating condition. Similarly, Torvela et al. (2014) detected UFP mainly composed of ash material in the emissions of a research biomass grate combustion unit (40 kW) combusting wood chips. Longhin et al. (2016) observed K, Mn and Ca in the UFP from a modern pellet boiler (25 kW). Park et al. (2013) analyzed water soluble components of PM from the combustion of different biomass materials (agricultural and forest residues) and found that the analyzed water soluble ions (e.g., organic, potassium, sulfate, chloride) showed higher mass in the UFP fraction than the higher particle diameters.

Existing research efforts for providing emission factors for chemical species of the particles from biomass combustion are mostly directed to characterize coarse ($d_p < 10 \mu\text{m}$), fine ($d_p < 2.5 \mu\text{m}$) and submicron particle emissions (e.g., Vicente et al., 2015; Lamberg et al., 2011 and many others). Differently from the

above mentioned studies our study focuses on the characterization of ultrafine particles (UFP, diameter $< 100 \text{ nm}$) that are indicated in recent research to present a series of specific characteristics and reactivity patterns that differ from that of larger particles (Diaz-Robles et al., 2014). Cassee et al. (2013) report in their review the concern for particle sizes such as ultrafine particles and certain chemical-specific constituents of PM such as sulfates, transition metals and polycyclic aromatic hydrocarbons without a definite conclusion on the toxicity of the PM released from biomass combustion. They reaffirm the need for further research to establish whether exposure to ultrafine particles alone can substantially contribute to the adverse effects of PM.

The present work is a part of a larger project (TOBICUP - TOXicity of Biomass COMbustion generated Ultrafine Particles) designed to gain deeper insight on the possible negative toxicological effects of UFP. TOBICUP project aims to verify the toxicological responses of the samples collected both directly from residential wood combustion emissions under burning cycles reflecting real-life situations and in ambient air at a sampling site where biomass burning for residential heating is widely used. Given the possible dependence of toxicological responses on the physical-chemical properties of the particles, the main goal of this study is the physical-chemical characterization of UFP from biomass burning in small-scale appliances.

Our previous experience has shown the importance of capturing the emission characteristics in the proximity of the source when the flue gas exiting the chimney is not fully diluted and the particulate matter emissions have not yet completely undergone oxidation by reactive atmospheric species (Ozgen et al., 2013). Taking into account the fact that the limited emission height of domestic heating appliances may increase the exposure of the population living nearby the source, the current study is designed also to reflect, as close as possible, the changes in the emissions, enhancing the dynamic processes involving the particles, exhausting the potential of the flue gas to condense upon further cooling.

2. Experimental section

2.1. Combustion appliances and test fuels

Two commercial residential solid biomass room heaters (one automatically stoked and one manually fed appliance) were tested in the Laboratory for Energy and Environment of Piacenza, Italy (LEAP) facility (www.leap.polimi.it). Detailed information on the test fuels is provided in the supplementary material (Table SM1).

Eight tests were performed with an 11.1 kW top-feed pellet stove. Pellets are fed from the internal storage to the burner pot. The ignition occurs by means of an electric device. The primary air is supplied from the bottom grate under the fuel bed and through the holes on vertical walls in correspondence of the lowest part of the flame zone. The secondary air (i.e., window flush) enters through slits in the upper part of the front window. The air and fuel supply rates are regulated automatically based on the stove heat

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