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Research article

# Wood washing: Influence on gaseous and particulate emissions during wood combustion in a domestic pellet stove



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# ABSTRACT

Nowadays, the use of biomass increasingly replaces the fossil fuels for the domestic heating production. In order to reduce pollutant emissions from biomass combustion, wood was washed at room temperature in order to represent natural rain leaching before burning in a recent pellet stove (2010s) of nominal output of 6.3 kW. Raw and washed woods were combusted for three different types of wood (oak, beech and fir) and the study focused on their particulate and gaseous emissions (Total Suspended Particles (TSP), Particulate Matter with diameter below 2.5  $\mu$ m (PM<sub>2.5</sub>), carbon monoxide (CO), nitrogen oxides (NOx) and Total Volatile Organic Compounds (TVOC)). Polycyclic Aromatic Hydrocarbons (PAH), aldehydes and wood tracers as phenols compounds were also measured. In addition, considering the toxic equivalent factor, the human health impact of adsorbed and gaseous PAH is considerably reduced (96%) in the case of washed fir combustion. Emission factors of CO and TSP for washed wood combustion also show a decrease up to 50% depending on the type of wood used. Furthermore, phenolic compounds, Benzene, Toluene, Ethylbenzene, Xylenes and Trimethylbenzene (BTEXT) emissions can also be reduced by the washing of biomass. Washed oak combustion leads to a clear decrease by 60% of the total of BTEXT. In the case of phenols emissions, phenol shows a significant decrease by 91% during the combustion of washed fir wood.

# 1. Introduction

Wood pellets have become an important fuel in domestic heat generation, since the costs of fossil fuels are rising and the emissions are nearly  $CO_2$  neutral. All households are incited to turn to the use of biomass as energy source for domestic heating. In contrast to other wood based fuels, the utilization of pellets is easy and automatic feeding to stoves and boiler is possible. Furthermore, the pelletization densifies the wood and produces a fuel with high energetic density [1]. On the one hand, wood pellets are a convenient choice for domestic fuel because of its simplicity of implementation and its low cost [2]. But on the other hand, wood pellets combustion, and more generally wood combustion, is a source of fine particles ( $PM_{2.5}$ ) and gaseous compounds as carbon monoxide (CO) or other incomplete combustion gas as nitrogen oxides (NOx) or Non-Methane Volatile Organic Compounds (NMVOC) [3–8]. But most emissions from pellets combustion are lower than wood log combustion [3,9] because in the case of wood logs, parameters as wood species, humidity

could disfavor the combustion process and generate big amounts of incomplete combustion products. As example, pollutant emissions from wood log combustion in domestic devices (open and closed fireplaces, traditional and advanced stoves) range from 20 and 120 g·kg<sup>-1</sup> for CO, are close to  $1 \text{ gkg}^{-1}$  for NOx, range from 2 to  $20 \text{ gkg}^{-1}$  for NMVOC and range from 0.2 to  $2 \text{ g/kg}^{-1}$  for PM emissions [3–6]. The high magnitude of emissions from small domestic devices well depends on several parameters as fuel quality and operating conditions (wall material of the combustion chamber, natural draft, primary and/or secondary air, operational practices, etc.). Whereas values from pellets combustion range from 1 to  $5 \text{ g/kg}^{-1}$  for CO, are close to  $1 \text{ g/kg}^{-1}$  for NOx, range from 60 to  $100 \text{ mg} \text{kg}^{-1}$  for NMVOC and range from 0.2 to 0.5 g kg<sup>-1</sup> for PM emissions [3,7,8]. More generally, combustion of wood pellets, due to a better complete combustion, is clearly less emissive in comparison with wood logs combustion. The automatic feed of the fuel and the possible presence of a lambda probe are advanced technologies that contribute to complete combustion [7,8].

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In the last decade, numerous studies were devoted to the evaluation of emission factors for gaseous compounds or particulate matter from residential wood combustion appliances [3,8,10]. The main gaseous pollutants usually followed from wood combustion are carbon monoxide, Volatile Organic Compounds (VOCs) [8]. Trace pollutants as Polycyclic Aromatic Hydrocarbons, benzene, toluene, ethylbenzene, xylenes, trimethylbenzene (BTEXT), phenols, aldehydes and ketones are often analyzed too [3,10]. Phenol and methoxy-phenols as syringol and guaiacol are also used as tracers of wood combustions [11]. Regarding particulate emissions, Total Suspended Particles (TSP) and PM<sub>2.5</sub> are constantly measured because it is well known that wood burning is one of the most emitting sources of fine particles [3,9,12].

Nowadays, several techniques are developed in order to reduce pollutants from wood combustion. Primary and secondary technologies must be distinguished: on the one hand, primary solutions focus on fuel quality and the conception of stoves and boilers, on the other hand, secondary solutions focus on the post-treatment of fumes [13-15]. Secondary technologies largely dominate using technologies as electrostatic filter, catalytic filter, or cyclones [16,17]. Secondary technologies aim principally at reducing particles emissions, but some techniques as catalytic filter allow to reduce gaseous emissions as carbon monoxide and VOCs [17]. Concerning primary techniques, important efforts from the 2000s were done in order to complain with different National and European standards [18-21] in terms of conception of stoves and boilers [22], choice of wood species and optimization of the air/fuel ratio [23]. One of the primary solutions to reduce pollutant emissions from biomass combustion could be a washing pretreatment of wood. Indeed, by washing wood, some compounds as minerals or extractives from wood could be removed. Deng et al. [24] have studied demineralization of some biomasses by a washing process with deionized water at temperatures ranging from 30 °C to 90 °C and results show that minerals as potassium or calcium could be extracted. For example from rice straw, extraction efficiency reaches respectively 87 and 19% at 30 °C for both elements. Jiang et al. [25] have also studied rice straw demineralization by several solvents as deionized water, chloride acid or phosphoric acid for example. Results show that, depending on the solvent, demineralization efficiency varies: calcium can be removed by 17% using deionized water versus 98% using chloride acid. Some extractives as phenols or carboxylic acids are also removed by wood washing [25-27].

Most of the studies have attempted to compare pollutant emissions as a function of the nature of the wood or the technology of the domestic heating appliances [22,23]. Main literature data focused on the influence of demineralization process on devolatilization phases of lignocellulosic polymers during thermogravimetric analysis [24,25,28]. Investigations on the influence of the wood preparation, particularly the washing process on gaseous and particulate pollutants emitted in the exhaust are scarce at real domestic scale [28]. This study aims at studying the leaching at room temperature of woods and their combustion (raw and washed biomasses) in a pellet stove to observe variations of gaseous and particulate emissions thanks to demineralization of wood. This work is a preliminary study of a French project supported by the National Agency of Energy and Environment (ADEME). The ultimate goal of this project is to study the influence of the natural leaching by rain of wood logs stored outside in forest for several months to a few years on gaseous and particulate pollutants during combustion in domestic devices as inserts and stoves. In order to get free of the variability due to the wood log fuel, it was decided to study the influence of leaching at laboratory scale using a pellet stove. Wood chips were then washed using demineralized water before preparing wood pellets. The stabilized conditions of a pellet burning system allow the detection of even small differences between different fuel types. This preliminary study on pellets washing aims to really understand and clearly explain the role of leaching on wood combustion with respect to gaseous and particulate emissions. Combustion of natural leached wood logs in a domestic stove will follow in a near future in order to compare these results.

#### 2. Experimentals

#### 2.1. Washing and pelletization protocols

Washing and pelletization protocols of biomasses are schematized in Fig. 1. Pre-treatment of beech, oak and fir chips was carried out at laboratory in a pilot according to the standard leaching protocol EN 12457/ 2 of May 2002 [29]. A quantity of 20 kg of dried wood chips was introduced into the pilot filled with 2001 of deionized water and mechanically shaken for 6 h. This operation was repeated three times but without drying wood samples between each cycle. For each cycle, the washing water is analyzed by ICP/OES (Thermo Scientific model ICAP 6300 DUO) for the quantification of minerals. The Total Organic Carbon (TOC) of residual leached solutions for 24 h was determined by catalytic oxidation of the organic carbon into dioxide carbon using a SHIMADZU TOC-VCSN apparatus. The moisture content of all raw and washed biomasses was brought to about 12 wt% (wet basis). Biomass chips were then ground and pelletized by a pellet press KAHL 14/175 equipped with a die having channels of 6 mm diameter and 22 or 26 mm length. Natural wood pellets with diameter close to 6 mm (purchased from SOFAG company, Arc sous Cicon, France) according to the DIN CERTCO standard were used as a reference during combustion tests [30].

## 2.2. Characterization of wood pellet samples

According to Table 1, the weight fractions of the different elements are in the same order of magnitude than those found for raw biomasses and lignocellulosic materials in literature [31]. The leaching process does not really affect elemental values CHONS. Both raw and washed woods show values having the same order of magnitude. O/C and H/C atomic ratio values are close to 0.7 and 1.5 respectively, according to Van Krevelen diagram defining biomasses [32-34]. A slightly decrease of the O/C ratios is observed for washed fir and oak, due to the dissolution of labile oxygen organic molecules as extractives (phenols, carboxylic acids, simple sugars, glycosides, fats, etc.) during the washing procedure [25,27]. This loss of organic molecules impacts the LHV as seen in Table 1. The leaching process also extracts low amounts of TOC as shown in Table 1. Oak is well-known to be a hard wood specie that contains higher amounts of tannins [35]. Biochemical composition of wood pellets was determined according the Van Soest's protocol [36] and results are given in Table 1 and are discussed in the following text (part 3).

Washing or leaching proceeds to demineralization of the wood with very high removal efficiencies of some minerals as potassium, sodium, calcium, sulfur and phosphor [24,28]. Raw and washed biomasses have been analyzed by ICP/OES (Thermo Scientific model ICAP 6300 DUO). Table 2 shows values of 10 main minerals contained in the raw biomasses and the percentage of extraction thereof from the washing process. Excepted for calcium removal efficiencies, values of the percentage of extraction are according to the literature [24,25,28,37], main soluble elements being K, Na and P with extraction balances ranging from 50 to 100% and depending of several factors as temperature, duration time and mass/volume ratios. Ratios of extraction for calcium drastically depend of its chemical speciation in the wood related to nature and the solubility of each salt (NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, CO<sub>3</sub><sup>2-</sup>, PO<sub>4</sub><sup>3-</sup>, etc.) [38].

# 2.3. Combustion tests

### 2.3.1. Experimental set-up platform

The experimental combustion setup is schematized in the Fig. 2. Combustions tests were performed in a pellet stove supplied by Hoben (Model H5 Signature Color Steel), complying with the European standard NF EN 14785 [20]. The power can be adjusted by an Intelligent Regulation System (SRI) from 1.3 kW to 6.3 kW. The steel inner wall of the combustion chamber is covered by vermiculite plates in order to

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