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Full Length Article

## Emissions of polycyclic aromatic hydrocarbons during biomass combustion in a drop tube furnace

J.M. Colom-Díaz<sup>a,\*</sup>, María U. Alzueta<sup>a</sup>, Ulisses Fernandes<sup>b</sup>, Mário Costa<sup>b</sup><sup>a</sup> Aragón Institute of Engineering Research (I3A), Department of Chemical and Environmental Engineering, University of Zaragoza, C/Mariano Esquillor s/n, 50018 Zaragoza, Spain<sup>b</sup> IDMEC, Mechanical Engineering Department, Instituto Superior Técnico, Universidade de Lisboa, Lisboa, Portugal

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## ABSTRACT

The objective of this work is to investigate experimentally the formation of polycyclic aromatic hydrocarbons (PAH) during the combustion of biomass in a drop tube furnace (DTF). A number of biomass fuels, including furniture residues, grape pomace, kiwi residues, olive residues, wheat straw, rice husk and platanus residues were used in this work, with the tests performed at three temperatures (900, 1000 and 1100 °C). The solid fuels feed rate was 23 g/h and the total air flow rate was 4 L/min, ensuring a residence time in the DTF of around 2 s. In order to collect the PAH in the effluent gas, a narrow tube containing XAD-2 resin was connected to the flue gas duct of the DTF. A quartz fiber filter was placed just before it to collect the particulate matter, including soot, present in the flue gas. The analysis and quantification of the PAH combined Soxhlet extraction and gas chromatograph-mass spectrometer. Flue gas concentrations of O<sub>2</sub>, CO<sub>2</sub>, CO, hydrocarbons and NO<sub>x</sub> were measured to gather information regarding the combustion conditions. The results showed two distinct features for the variation of the total PAH emissions: one decreasing with temperature and other with a maximum at 1000 °C. Grape pomace, kiwi residues and platanus residues presented the lowest PAHs emission (20.8–54.2 mg PAH/kg fuel). A direct relation between the total amount of PAHs and the toxic equivalency value was found.

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### 1. Introduction

Biomass is a well-known fuel that has become important as an energy carrier in the last decades due to its high diversity and availability. Biomass can be seen as a fuel originated from organic materials such as plants, leftovers from agricultural materials and forestry processes, as well as organic industrial, animal and human wastes. Nowadays, most biomass combustion processes are optimized for woody biomass [1,2]. However, due to economic and environmental reasons, only a part of the available forest biomass can be used. Therefore, in addition to forest biomass, it is critical to use also agricultural residues for energy purposes. The extreme variability of the biomass feedstock demands for an extensive investigation on the impact of its composition and characteristics in their combustion behaviour.

Biomass, specially wood, are considered to be a major emission source of local ambient pollution [1,2], namely particulate matter (PM), soot and polycyclic aromatic hydrocarbons (PAH), which are all a great concern and associated to highly carcinogenic and muta-

genic compounds [3,4]. The USA Environmental Protection Agency (EPA) released a list with 16 priority compounds, known as 16 EPA-PAH, based on their potential toxicity for human exposure or frequency of occurrence at hazardous waste sites. A number of studies have focused on this topic, reporting emissions of PAH from biomass combustion, specially wood combustion [5–7]. Wood combustion originates more emissions of PAHs than any other source commonly used for energy purposes [8]. The appliance used (fireplace versus wood stove) and the type of wood (hardwood, softwood or synthetic log) have been reported to have the largest effect on the composition of the emissions from wood combustion [9,10]. Studies on emissions from residential biomass boilers and stove systems using non-wood fuels are rare. An exception is the work of Chandrasekaran et al. [11], who measured the emissions from a residential-scale biomass boiler fired with pellets of wood, grass and a blend of grass and corn. They found that grass pellets can not be used as a fuel in typical wood pellet systems due to its high ash and chlorine content, and higher PAHs emission compared to those from the wood pellets firing.

The technologies for the mitigation of PM emissions from domestic boilers were reviewed by Lim et al. [12], and gas sampling methods for residential biomass fired appliances were

\* Corresponding author.

E-mail address: [juanmcd@unizar.es](mailto:juanmcd@unizar.es) (J.M. Colom-Díaz).

discussed by Boman and coworkers [13]. The use of biomass has recently risen to 13–14% of the primary energy consumption in the world, together with an increase of modern pellet stoves and pellet consumption [14]. In Europe, the pellet market is growing in small- and medium-scale heat production units as well as in large-scale power generation plants, which can contribute to fulfill the renewable heat and electricity targets set by the European Union [14]. In this context, there is an urgent need for detailed characterization and quantification of the emissions under controlled and standardized conditions, using different fuels and combustion techniques [13].

Since the operating conditions may play a significant role in the emissions from biomass combustion [15], the use of a drop tube furnace (DTF), under well controlled conditions, can help to bring new information on the formation and emission of PAH. DTFs simulate well real combustion processes under controlled conditions, namely excess air, residence time, heating rate and temperature, while allowing for the collection of detailed and reliable data. To the best of our knowledge, there are no studies in the literature related to PAHs from the combustion of biomass in DTFs. Chen et al. [16] studied the emission characteristics of PAHs during coal and sewage sludge co-combustion in a DTF, concluding that the PAHs concentration in the flue gas exhibited a maximum as a function of the temperature in the 950–1250 °C range. Panagiotou et al. [17] studied the effect of the equivalence ratio on PAHs emission from the combustion/pyrolysis of polyvinyl chloride (PVC), polyethylene (PE), and polystyrene (PS) particles in a DTF, finding that for all polymers the total amount of PAHs increased with increasing the equivalence ratio and that the highest amount of PAHs were emitted under pyrolytic conditions in N<sub>2</sub> at varying particle loadings. In this context, the present work evaluates the formation of the 16 priority PAH during the combustion of 7 different biomass residues in a DTF.

## 2. Materials and methods

### 2.1. Biomass fuels

The biomass fuels used in this study were furniture residues, grape pomace, kiwi residues, olive residues, wheat straw, rice husk

**Table 1**  
Characteristics of the biomass residues.

Parameter	Furniture residues	Grape pomace	Kiwi residues	Olive residues	Wheat straw	Rice husk	Platanus residues
Proximate analysis (wt.%, as received)							
Volatiles	77.2	48.4	71.7	72.6	63.8	65.0	73.5
Fixed Carbon	16.3	18.6	17.6	16.2	14.9	13.3	16.0
Moisture	5.7	30.2	8.3	8.7	8.3	9.5	9.3
Ash	0.8	2.8	2.4	2.5	13.0	12.2	1.2
Ultimate analysis (wt.%, dry ash free)							
Carbon	57.1	51.1	48.2	47.9	42.0	44.0	47.6
Hydrogen	7.3	6.7	6.4	6.2	5.4	5.6	6.0
Nitrogen	3.8	1.9	0.7	0.7	0.6	0.6	0.5
Sulphur	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Oxygen	31.6	40.1	44.5	45.0	51.8	49.6	45.7
Ash analysis (wt.%, dry basis)							
SiO <sub>2</sub>	16.0	5.5	1.5	3.3	42.5	90.4	1.3
Al <sub>2</sub> O <sub>3</sub>	11.6	1.0	1.5	2.2	8.5	1.0	0.8
Fe <sub>2</sub> O <sub>3</sub>	5.8	1.2	0.8	1.3	4.9	0.4	0.6
CaO	36.2	37.8	44.7	35.9	26.9	1.6	59.9
SO <sub>3</sub>	3.9	1.7	2.5	1.1	1.1	0.3	2.7
MgO	4.4	7.2	5.8	4.8	4.0	0.9	8.5
P <sub>2</sub> O <sub>5</sub>	2.6	19.7	15.8	18.3	2.9	1.1	12.9
K <sub>2</sub> O	6.6	24.7	23.2	28.1	7.2	2.5	8.7
Na <sub>2</sub> O	2.9	0.4	1.1	1.8	0.7	0.3	2.7
Cl	0.1	<0.01	0.2	0.6	0.6	0.5	1.0
Other oxides	9.9	0.8	2.9	2.6	0.7	1	0.9
Lower heating value (MJ/kg)	16.6	19.8	17.9	18.0	13.2	14.5	18.1

and platanus residues. Table 1 shows the properties of the biomass fuels, including the proximate and ultimate analysis, the ash composition and the low heating value.

### 2.2. Drop tube furnace and test conditions

The present study was performed in the DTF shown in Fig. 1, and described in detail elsewhere [18]. Briefly, the DTF is electrically heated and operates at a maximum temperature of 1100 °C. The combustion chamber is a cylindrical ceramic tube with an inner diameter of 38 mm and a length of 1300 mm. The furnace wall temperatures are continuously monitored by eight type-K thermocouples uniformly distributed along the combustion chamber. A water-cooled injector, placed at the top end of the DTF, was used to feed the solid fuels and the air to the combustion chamber. A twin screw volumetric feeder transfers the solid fuels to an ejector system from which the particles are air-transported to the water-cooled injector.

In this study, measurements were carried out for all biomass fuels for DTF wall temperatures of 900 °C, 1000 °C and 1100 °C. The feeding rate of the solid fuels was 23 g/h and the total air flow rate was 4 L/min, ensuring a residence time in the DTF of around 2 s. The PAH collection started once the DTF reached steady-state conditions, with each measurement lasting for about 15 min.

### 2.3. Experimental techniques

Flue gas concentrations of O<sub>2</sub>, CO<sub>2</sub>, CO, hydrocarbons (HC) and NO<sub>x</sub> were measured to gather information regarding the combustion conditions. These measurements were obtained with the aid of a 1.2 m long, water-cooled stainless-steel probe. The gas sample was drawn through the probe and part of the system by an oil-free diaphragm pump. A condenser removed the main particulate burden and condensate. A filter and two driers removed any residual particles and moisture so that a constant supply of clean dry combustion gases was delivered to the analyzers through a manifold to give species concentration on a dry basis. The analytical instrumentation included a magnetic pressure analyzer for O<sub>2</sub> concentration measurements, a non-dispersive infrared gas analyzer for CO<sub>2</sub> and CO concentration measurements, a flame ionization detector

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