

Gaseous and particulate emissions from the combustion of wood in a Portuguese stove

Márcio Duarte*, Estela Vicente*, Ana Calvo**, Teresa Nunes*, Luís Tarelho*, Célia Alves*

* Centre for Environmental and Marine Studies, Department of Environment, University of Aveiro, 3800-193 Aveiro, Portugal

** Department of Physics, IMARENAB University of León, 24071 León, Spain

Email: marcioduarte@ua.pt

ABSTRACT

In Europe, biomass burning is a source of several pollutants, especially in winter time. Several human health problems have been associated with pollutants released from biomass burning. The objective of this work was to characterize the emissions of a wide range of pollutants from a typical Portuguese wood stove fuelled with two common species: eucalypt and pine. The highest emissions for several gaseous compounds analysed, excepting hexane, were obtained for eucalypt. Pine presented the highest emission of PM₁₀.

Keywords: Biomass combustion, emission factors, gaseous emissions, PM₁₀

1. INTRODUCTION

The importance of biomass fuels has increased, in recent years, mainly for two reasons: (i) the effort to control emissions of greenhouse gases from fossil fuels, (ii) their CO₂-neutrality and the fact that they represent renewable energy sources [1], and (iii) the increase costs associated with fossil fuels. Despite this, biomass combustion is recognized as a major source of air pollutants with potential to cause health hazards [2] and with impacts on air quality, as it has been proved in several studies [3–5]. Biomass burning releases considerable amounts of airborne pollutants, such as nitrogen oxides, methane and volatile organic compounds (VOCs), polyaromatic hydrocarbons, furans and dioxins, but also particulate matter, which has a major impact on atmospheric pollution, especially in rural sites in Europe during winter time [6].

As in many other countries, biomass burning is a common practice in Portugal, where it is mainly used for domestic heating or cooking. Estimates of annual wood consumption are 1950 kton in domestic appliances [7].

The lack of information regarding the emission composition and emission factors (EF) for biomass burning in Portugal, leads to higher errors when estimating emissions. This is due to the use of EFs from US, Alpine and Scandinavian biofuels, which, in general, are uncommon in southern Europe [6, 8, 9]. Besides the differences in the fuel composition, the design and characteristics of the appliances are known to influence the emissions from biomass burning [10, 11].

In order to improve estimates of the amount and profiles of gaseous and particle emissions, it is necessary to burn biomass fuels commonly used in Portugal, as well as using the same type of appliances.

This study aims to characterize the particle and gaseous emissions from Portuguese wooden biofuels burned in a typical woodstove.

2. EXPERIMENT

2.1 Fuel characteristics

The biomass fuels selected for this study were pine (*Pinus pinaster*) and eucalypt (*Eucalyptus globulus*), two types of wood commonly used in Portugal (Table 1). The logs were cut into pieces of 0.3 to 0.4 m in length. In each combustion cycle, 1.7 to 2.0 kg of biomass was burned. The combustion of each batch of fuel lasted between 45 and 60 minutes.

Table 1 - Chemical characteristics of the biofuels (bdl – below detection limit).

		Pine	Eucalypt
Proximate analysis (% wt, as received)	Moisture	9.6	14.8
	Ash	0.46	0.25
Ultimate analysis (% wt, dry basis)	C	51.4	48.6
	H	6.2	6.2
	N	0.16	0.16
	S	bdl	bdl
	O (by difference)	41.78	44.79

2.2 Combustion experiments, gas and PM₁₀ sampling

The woodstove of this study is commonly used in Portugal for domestic heating (air heating). It is an equipment produced by the Portuguese company Solzaima [12]. The stove is made in stainless steel and has a cast iron front panel and grate. The volume of the combustion chamber is around 0.09 m³, corresponding to 0.44 m height, 0.59 m width and 0.36 m depth. It is operated manually in batch mode with handheld control of combustion air. The combustion air enters the combustion chamber throughout a cylindrical structure/tube of 6 cm in diameter, located beneath the door, and flows throughout the grate and biomass fixed bed. A mass flow meter (Kurz, Model: 500-2.0-P 40) measures the air flow rate entering the combustion chamber. Furthermore, this cylindrical device supports the hopper for collecting the bottom ashes, located below the grate of the stove. The driving force for the air entering the combustion chamber is natural convection resulting from the up flowing column of hot combustion flue gases throughout the vertical chimney. The wood stove heats the room air by a combination of radiation, natural and forced convection. The stove is equipped with a vertical exhaust duct (chimney) with 0.20 m internal diameter and 3.3 m height; the exhaust duct was thermally insulated (with cerablanket, 128 kg m⁻³, 0.025 m,

0.23 W m⁻¹ K⁻¹) in the first 1 m above the stove exit, due to operational safety reasons.

A fixed grate supports the logs under combustion. A weight sensor (DSEUROPE Model 535QD-A5, C Figure 7) was connected to the grate in the stove, with the objective of continuous monitoring the weight of fuel in the burning fixed bed. However, the stove is not completely tight and some air can enter without being measured. To face this problem a Pitot coupled to a pressure sensor (JUMO, model Typ 4304) and a type-K thermocouple are used to determine the flow throughout the chimney and the total amount of air entering the combustion chamber.

The flue gas sampling and characterization was made using a Gaset CEMS FTIR measuring system (FTIR, Figure 7), which is designed for continuous emission monitoring measurements. The gases analysed were H₂O, CO₂, CO, N₂O, NO, NO₂, SO₂, HCl, HF, NH₃, CH₄, C₂H₆, C₃H₈, C₂H₄, C₆H₁₄ and CHO. The system consists in a FTIR gas analyser, a Gaset industrial computer and a Gaset sampling system. All parts of the system are 19" rack mounted and are installed on the pull-out shelves. The Gaset CEMS includes all power connections and temperature controllers for heated lines and heated sample probe. The operation of the system is fully automatic and controlled by the Calcmet software [13].

For O₂ monitoring (O₂, Figure 7), the gas sampling and characterization system included a water-cooled sampling probe, a set of gas conditioning and distribution units, and an on-line gas analyser, ADC 700 (paramagnetic) with a Servomex module. The combustion flue gas was sampled at a flow rate of 2 L.min⁻¹ (atmospheric pressure and temperature). The operation, monitoring, conditioning and characterization were performed by a computer based control and data acquisition hardware and software system.

Temperature in the combustion chamber and in different locations along the system was monitored with K-type thermocouples.

Particulate matter <10 µm (PM₁₀) in the combustion flue gas were collected in a dilution tunnel located downstream of the chimney (Q, Figure 7). This dilution tunnel (O, Figure 7) consists of a tube of circular section with 11 m length and 0.20 m internal diameter. The gas velocity in the cross section of the dilution tunnel was determined using a Pitot tube (Testo AG 808; P, Figure 7), a pressure sensor and a K-type thermocouple; this allowed the calculation of the volumetric gas flow rate throughout the tunnel, and respective combustion gas dilution ratio. Particulate sampling points were located at ~10 m downstream the dilution tunnel entrance. The aim of this dilution source sampling is to simulate the rapid cooling and dilution that occurs when exhaust mixes with the atmospheric air. Gas-particle partitioning of semi volatile material in the combustion flue gas will be influenced by these processes [14].

PM₁₀ was collected on quartz filters (47 mm diameter) using a low volume sampler. The sampling train included a specific sampling head (PM₁₀), a pump operating at 38 L min⁻¹ (at atmospheric pressure and temperature), and a control and data storage unit, all making part of a TCR, TECORA (model 2.004.01) instrument (R and Q, Figure 7). A K-type thermocouple was placed near the sampling head to characterize the flue gas (combustion flue gas from the stove diluted with atmospheric air) temperature. In order to maintain the sampling flow rate and to avoid pressure drop across the filter media, the filter was replaced several times during the combustion cycle; each filter replacement lasted less than one minute [15].

3. RESULTS AND DISCUSSION

3.1 Concentration profiles

During the combustion tests with both fuels, it was observed that the FTIR analyser has high uncertainty on measurements in the initial stage of the combustion process, i.e. during the initial period of wood heating and devolatilisation. This problem seems to be caused by the high concentrations of water in the flue gas, which is released during the first stage of the combustion process. This problem is flagged by the analyser software and corresponds to high residual values in spectra analysis. However, it does not affect all the gases in the same way and some of them are not affected at all. Due to this, there is a lack of data in the initial stage of the combustion process for some gases (e.g., CO, CO₂, CH₄, NO, SO₂, NH₃, HCl).

3.1.1 Pine

During the combustion of pine, the highest emissions for CO₂ were coincident with those of H₂O. The lowest O₂ concentrations were concomitantly registered (Figure 1). On the other hand, carbon monoxide (CO) concentrations decreased during this period to values identical to those of the beginning. The same occurs with some hydrocarbons (Figure 3). After that, CO achieved an even lower value, due to the high temperatures in the combustion chamber and the high O₂ concentrations, which enabled an extended conversion of unburned material to CO₂ and H₂O.

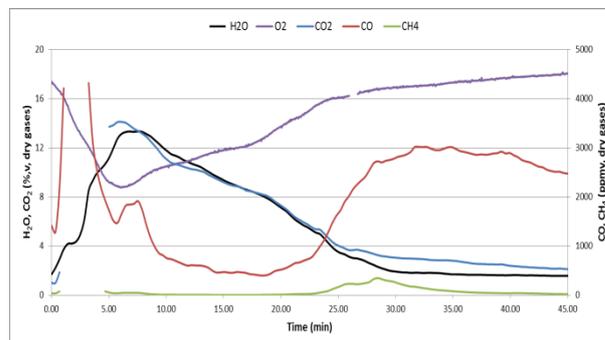


Figure 1 - H₂O, O₂, CO₂, CO and CH₄ emission profiles during the combustion of pine.

Ammonia presented a similar profile to that of CO. However, concentrations were lower and in the first 5 minutes it wasn't detected (Figure 2). Despite sulfur was below the detection limit in the fuel, it was present in the flue gas with high emission in beginning of the combustion. However, it seems that the higher water contents in the gas have a bigger effect in SO₂ than in other gases. SO₂ presented an average concentration of 8 ppm. NO₂ and N₂O had very low concentrations, 2.5 and 0.7 ppm, respectively, while, on the other hand, NO showed a similar emission profile to those of CO₂ and water vapor, with an average concentration of 30 ppm.

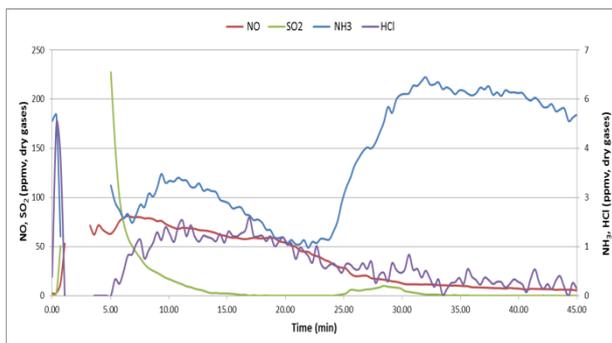


Figure 2 – NO, SO₂, NH₃ and HCl emission profiles during combustion of pine.

Figure 3 depicts the evolution of several hydrocarbons throughout the combustion experiments. Methane presented the highest average concentration among all (60 ppm). However, in the first 5 minutes, CH₄ concentrations were surpassed by CHO_H. Despite the values were ignored, it appears that the lower temperatures in the early stages of combustion promote the emission of formaldehyde and ethane.

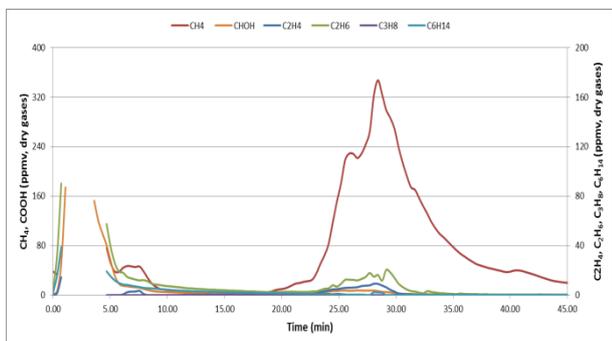


Figure 3 – CH₄, CHO_H, C₂H₄, C₂H₆, C₃H₈ and C₆H₁₄ emission profiles during the combustion of pine.

3.1.2 Eucalypt

The combustion of eucalypt showed, in general, a similar behaviour to that of pine, despite the higher concentrations of some gases.

During the heating and devolatilisation phases, CO reached concentrations of 7000 ppm. As observed in the case of pine, these levels were registered with high water content, which leads to high uncertainty in the measuring method, and thus those values are not shown. Also, CO₂ and H₂O presented similar profiles with average concentrations of 6.4% and 6.1%, respectively.

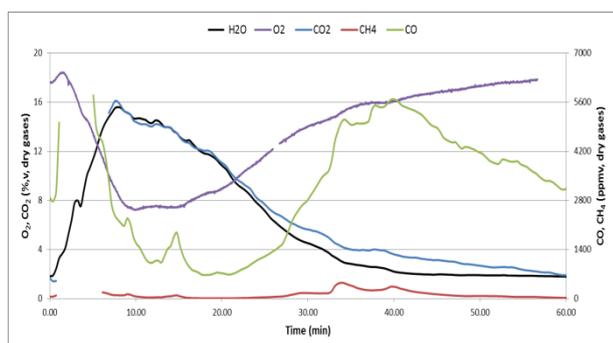


Figure 4 - H₂O, O₂, CO₂, CO and CH₄ emission profiles during the combustion of eucalypt.

Figure 5 shows the evolution of NO, SO₂, NH₃ and HCl concentrations in the exit flue gases. It is observed that SO₂ has a similar behaviour to that of pine. The flue gas concentrations of NO₂ and N₂O were lower than 3 and 1.5 ppm, respectively. The mean nitrogen monoxide concentration was 35 ppm, maximizing at 85 ppm. These values are very close to those registered for pine combustion (30 ppm average and 81 ppm max). The combustion of eucalypt has a higher emission of HCl than pine, registering an average value of 10 ppm and a maximum of 31 ppm, while the corresponding values for pine were 0.8 ppm and 4.8 ppm.

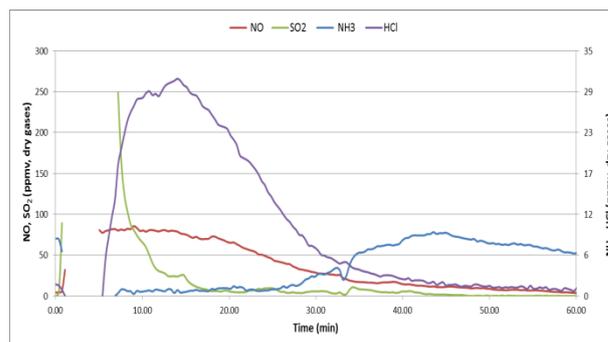


Figure 5 - NO, SO₂, NH₃ and HCl emission profiles during the combustion of eucalypt.

The emission profiles of various hydrocarbons is similar (Figure 6), and in some extent comparable to those of pine. However, in the case of eucalypt, the concentrations are higher. Methane was emitted at an average concentration of 105 ppm, achieving a maximum value of 448 ppm. Formaldehyde presented an average concentration of 11 ppm, but peaked at 218 ppm in the initial stage of the combustion process. Another difference between pine and eucalypt occurs around 30 minutes in to combustion cycle. Despite the increasing O₂ concentration, all hydrocarbons (and CO) suffer an increase in their emission. In the combustion of pine this effect is not so marked as in the case of eucalypt, except for methane.

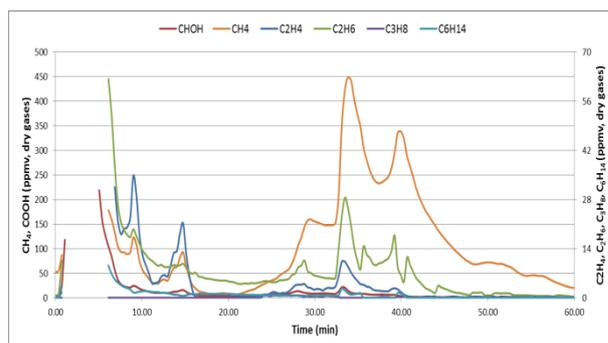


Figure 6 - CH₄, CHO_H, C₂H₄, C₂H₆, C₃H₈ and C₆H₁₄ emission profiles during the combustion of eucalypt.

3.2 Emission Factors

The emission factors calculated for the experiments carried out are shown in Table 2. Significant differences in the emission factors of eucalypt and pine are observed. Eucalypt presented the highest emission factors for all gaseous species (except C₆H₁₄), while pine exhibited the highest emission factor for PM₁₀. This result is opposite to what was expected, considering that low moisture content of the wood fuel (as the pine) can be traduced by higher combustion rates, creating zones with a deficit in O₂,

thus, enhancing the emission of several gaseous pollutants and also particulate matter emissions [16].

The EFs of CO, CH₄ and CHO_H were almost double for eucalypt when compared with pine. The differences observed between eucalypt and pine for NO (84.8 vs 68 mg·MJ⁻¹), SO₂ (95 vs 64 mg·MJ⁻¹) and HCl (16.6 vs 0.735 mg·MJ⁻¹) are relevant. These differences could be likely explained by distinct element uptakes by the trees due to growth under contrasting environmental conditions.

Table 2 - Emission factors for gases and PM₁₀. All emission factors are in dry basis and corrected to 13% O₂.

		Eucalypt		Pine	
		Average	std.	Average	std.
CO	[g·MJ ⁻¹]	6.5	0.6	3.8	0.5
N ₂ O		3.6	0.3	2.5	0.009
NO		85	0.2	68	5
NO ₂		16	5	12	2.1
SO ₂		95	29	64	24
NH ₃		4.4	1.5	4.5	0.95
HCl		17	2.8	0.73	0.39
HF	[mg·MJ ⁻¹]	0.03	0.027	0.001	0.001
CH ₄		155	12	88	10
C ₂ H ₆		12	2.0	8.5	0.95
C ₂ H ₄		6.6	2.2	7	5
C ₃ H ₈		0.76	0.07	0.19	0.18
C ₆ H ₁₄		5	0.1	11	2.5
CHOH		41	13	21	2.1
PM ₁₀		229	122	345	98

4. CONCLUSIONS

Particulate matter and gaseous emission factors and emission profiles along time were obtained for the residential combustion of two widespread Portuguese woods. The combustion experiments carried out showed important differences between the two types of biomass fuels. Overall, eucalypt presented the highest concentrations and EFs for of several gaseous pollutants, while pine produced the highest PM₁₀ EFs.

Despite the similarities between flue gas concentration profiles along time for pine and eucalypt, the amounts of several gaseous compounds emitted are higher for eucalypt, especially for SO₂, HCl and hydrocarbons.

ACKNOWLEDGMENT

This work was supported by the European Commission through the Life+2011 programme, project "AIRUSE - Testing and Development of Air Quality Mitigation Measures in Southern Europe" (LIFE 11/ENV/ES/000584), and

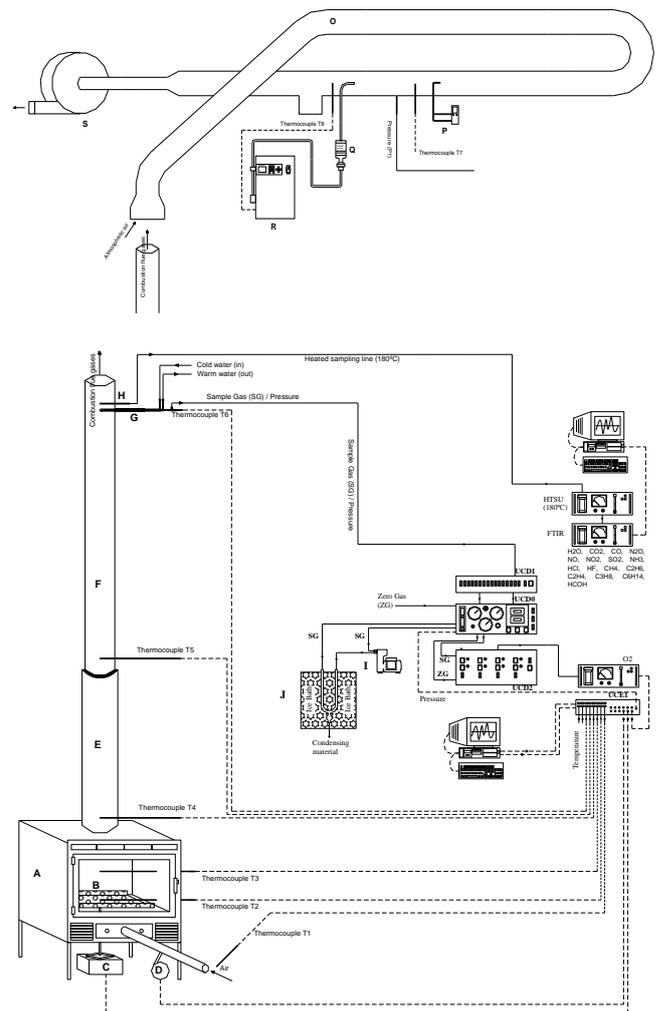


Figure 7 - Schematic representation of the experimental installation. Dashed line - Electric circuit; Continuous line - Pneumatic circuit. A - Stove, B - Grate of the stove, C - Load cell (weight sensor), D - Air flow meter, E - Thermal insulation of the exhaust duct, F - Exhaust duct (Chimney), G - Water-cooled gas sampling probe, H - Heated sampling line, UCD0, UCD1, UCD2 - Command and gas distribution units, I - Gas sampling pump, J - Gas condensation unit for moisture removal, O₂ - Automatic on-line gas analyzer, UCE1 - Electronic command unit, O - Dilution tunnel, W - Pitot tube, Q - Sampling head for PM₁₀ (TECORA), R - TECORA control and data acquisition system, S - Ventilator.

REFERENCES

- [1] PASTORELLO, Cinzia, Stefano CASERINI, Silvia GALANTE, Panagiota DILARA and Fabio GALLETTI. Importance of activity data for improving the residential wood combustion emission inventory at regional level. *Atmospheric Environment*. 2011, vol. 45, no. 17, pp. 2869–2876. ISSN 13522310. doi:10.1016/j.atmosenv.2011.02.070

- [2] KOCBACH BØLLING, Anette, Joakim PAGELS, Karl Espen YTTRI, Lars BARREGARD, Gerd SALLSTEN, Per E SCHWARZE and Christoffer BOMAN. Health effects of residential wood smoke particles: the importance of combustion conditions and physicochemical particle properties. *Particle and fibre toxicology*. 2009, vol. 6, no. i, p. 29. ISSN 1743-8977. doi:10.1186/1743-8977-6-29
- [3] HELLÉN, H, H HAKOLA, S HAAPARANTA, H PIETARILA and M KAUHANIEMI. Influence of residential wood combustion on local air quality. *The Science of the total environment* [online]. 2008, vol. 393, no. 2-3, pp. 283–90. ISSN 0048-9697. doi:10.1016/j.scitotenv.2008.01.019
- [4] GLASIUS, M, M KETZEL, P WAHLIN, B JENSEN, J MONSTER, R BERKOWICZ and F PALMGREN. Impact of wood combustion on particle levels in a residential area in Denmark. *Atmospheric Environment* [online]. 2006, vol. 40, no. 37, pp. 7115–7124. ISSN 13522310. doi:10.1016/j.atmosenv.2006.06.047
- [5] GIANELLE, Vorne, Cristina COLOMBI, Stefano CASERINI, Senem OZGEN, Silvia GALANTE, Alessandro MARONGIU and Guido LANZANI. Benzo(a)pyrene air concentrations and emission inventory in Lombardy region, Italy. *Atmospheric Pollution Research*. 2013, vol. 4, pp. 257–266. ISSN 13091042. doi:10.5094/APR.2013.028
- [6] JOHANSSON, Linda S, Bo LECKNER, Lennart GUSTAVSSON, David COOPER, Claes TULLIN and Annika POTTER. Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets. *Atmospheric Environment*. 2004, vol. 38, no. 25, pp. 4183–4195. ISSN 13522310. doi:10.1016/j.atmosenv.2004.04.020
- [7] GONÇALVES, Cátia, Célia ALVES and Casimiro PIO. Inventory of fine particulate organic compound emissions from residential wood combustion in Portugal. *Atmospheric Environment*. 2012, vol. 50, pp. 297–306. ISSN 13522310. doi:10.1016/j.atmosenv.2011.12.013
- [8] FINE, PM. Chemical characterization of fine particle emissions from the wood stove combustion of prevalent United States tree species. *Environmental Engineering Science*. 2004, vol. 21, no. 6. doi: 10.1089/ees.2004.21.705
- [9] DUARTE, M A C, A I CALVO, L A C TARELHO, T NUNES, M EVTYUGINA and C A ALVES. Carbonaceous compounds emitted from domestic biomass burning. European Aerosol Conference. 2012.
- [10] MCDONALD, Jacob D., Barbara ZIELINSKA, Eric M. FUJITA, John C. SAGEBIEL, Judith C. CHOW and John G. WATSON. Fine Particle and Gaseous Emission Rates from Residential Wood Combustion. *Environmental Science & Technology*. 2000, vol. 34, no. 11, pp. 2080–2091. ISSN 0013-936X. doi:10.1021/es9909632
- [11] JOHANSSON, L.S., C. TULLIN, B. LECKNER and P. SJÖVALL. Particle emissions from biomass combustion in small combustors. *Biomass and Bioenergy*. 2003, vol. 25, no. 4, pp. 435–446. March 2014]. ISSN 09619534. doi:10.1016/S0961-9534(03)00036-9
- [12] *Solzaima*: <http://www.solzaima.pt/>
- [13] *Gasmet*: <http://www.gasmet.com/>
- [14] GONÇALVES, Cátia, Célia ALVES, Margarita EVTYUGINA, Fátima MIRANTE, Casimiro PIO, Alexandre CASEIRO, Christoph SCHMIDL, Heidi BAUER and Fernando CARVALHO. Characterisation of PM10 emissions from woodstove combustion of common woods grown in Portugal. *Atmospheric Environment*. 2010, vol. 44, no. 35, pp. 4474–4480. ISSN 13522310. doi:10.1016/j.atmosenv.2010.07.026

[15] CALVO, AI and LAC TARELHO. Operating conditions of a Portuguese wood stove. 4th International Congress of Energy and Environment, Merida, Spain, 2011, p. 1–4.

http://www.cesam.ua.pt/subsites/files/calvo_et_al_1_1_.pdf

[16] SIMONEIT, Bernd R.T. *Biomass burning — a review of organic tracers for smoke from incomplete combustion*. 2002.

doi:10.1016/S0883-2927(01)00061-0