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Almond Shells and Exhausted Olive Cake as Fuels for Biomass Domestic Boilers: Optimization, Performance and Pollutant Emissions

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Abstract: The combustion of two non-woody types of biomass (almond shells and exhausted olive cake) in a domestic boiler at different loads was studied in order to evaluate their suitability as fuels. To select the optimal boiler operating conditions (excess air, primary/secondary air ratio and grate vibration), which allows for lower CO and particulate matter emissions for each biomass and load, a statistical design of experiments was performed. Similar optimal operating conditions were found for both fuels at nominal load (excess air: 1.5, primary/secondary air ratio: 20/80), the grate vibration being the only parameter to be modified due to the different ash content (45 and 20 s for almond shells and exhausted olive cake, respectively). At partial load, a slightly higher excess air (1.6) and a higher proportion of primary air (50/50) were needed in the case of almond shells. Results showed higher CO and lower NO_x and PM emissions at partial load for both fuels. The high ash content of exhausted olive cake deteriorated its combustion process (accumulated ashes were observed in the fireplace). Gaseous and solid emissions did not fulfil the UNE-EN 303-5 limits for any fuel or condition; although, almond shells seem to be a much more suitable fuel since they could be used just blended with a small quantity of a high-quality biomass or additive. However, exhausted olive cake not only led to a very poor efficiency at partial load (74%), clearly below the minimum required by the standard (77%), but also to an unacceptable pollutant emission level. So, this latter fuel would require a high blending ratio with another type of biomass, pre-treatments for reducing the alkali compounds and/or significant technological modifications allowing for a proper ash handling.

Keywords: agricultural fuels; small-scale combustion; air staging; gaseous emissions; particulate matter

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1. Introduction

The importance of biomass for producing thermal energy, both in the industry and the domestic sectors, is growing very quickly not only because of the corresponding reduction in the consumption of fossil fuels (and thus on the derived carbon dioxide (CO_2) emissions), but also as a way to guarantee the energy supply by using domestic fuels. The EU Renewable Energy Directive [1] establishes certain objectives for the year 2030, including that at least 32% of the final energy consumption should be provided by renewable sources (Spain reached 21.2% in 2020). Moreover, this directive encourages an annual increase of 1.3% in the use of renewable sources for thermal energy (heating and cooling sector) generation as well as a reduction in fossil fuels up to 80% for new installations in 2026. The Spanish National Energy and Climate Plan 2021–2030 [2] has established objectives aligned with this directive.

Biomass contributes more than 4% of the final energy consumption in Spain [3]. Regarding the residential sector, biomass combustion accounts for around 31% [4] in heating systems, wood pellets being the main commercial biofuel. Although wood-derived fuels are currently the main type of biomass, agri-food residues can be the key to supplying the growing expected demand since they are more sustainable and do not compete with other activities (paper and wood processing industry). As the main olive producer in the world (34.4% of the total world production) and the second of almonds (10.1%) [5], Spain generates a large amount of annual residue that is generally discarded or locally used in small cogeneration (CHP) plants in which the residue is generated (wine and olive oil industry). However, non-woody biomass can cause operating problems derived from its composition, the ash and alkali metals content being the main drawbacks. Emissions from biomass combustion are mainly produced by incomplete combustion (carbon monoxide (CO), hydrocarbons (C_xH_y), polycyclic aromatic hydrocarbons (PAH) and particulate matter (PM)), complete combustion (nitrogen oxides (NO_x), sulfur oxides (SO_x), hydrogen chloride (HCl) and PM) and ash formation (bottom and fly ash, and deposits) [6,7]. Due to their ash and alkali metal (such as potassium (K) and sodium (Na)) content as well as chlorine (Cl) and sulfur (S), the combustion of agro-food wastes is associated with high emissions of coarse fly-ashes (PM > 1 μm) and aerosols (PM < 1 μm) [7]. Pollutant emissions in residential applications are limited by UNE-EN 303-5 [8], UNE-EN 16510-1 [9] and Regulation (EU) 2015/1189 [10]. However, these standards were defined for high-quality solid fuels (such as those derived from wood), and they are very challenging to be fulfilled when using other fuels with residual origin. Thus, additional knowledge regarding the combustion behavior of these new and highly potential biofuels is required for optimizing the operating conditions of existing boilers, for designing new devices and for efficiently handling ashes.

To reduce the emissions and to optimize the combustion of domestic boilers, several authors [11–13] have investigated the effect of excess air and air staging as it is very cost-effective to improve the boiler performance. Excess air is necessary to facilitate complete combustion of the fuel and depends on the type of biomass [14]. It is common to use excess air between 1.5 and 2 to achieve complete combustion; although, lower CO emissions can also be achieved with a lower excess air through an appropriate air distribution [15]. The primary air flow rate must be sufficient to facilitate the volatilization of the fuel and the complete combustion of the carbonaceous residue, avoiding unburnt residues. Whereas the secondary air must promote mixing between the air and the flue gas to achieve complete combustion of the latter. In general, secondary air flow is favored over primary air flow. A very high primary air flow rate reduces the residence time in the primary combustion zone and increases particulate emission [16], as well as results in a lower combustion chamber temperature and lower efficiency [17]. If this flow rate is too low, the reaction rate is reduced and results in a lower combustion efficiency [17]. Khodaeli et al. [11] analyzed different air distribution strategies at lab scale by using wood pellets and they reported up to a 50% reduction in CO and a lower particulate matter concentration when a secondary air module was added at the top of the bed. Lamberg et al. [12] studied the combustion of wood pellets in a 25 kW boiler and observed that increasing the secondary air flow improved the mixing of gases in the secondary combustion zone and resulted in lower emissions of unburned gases and particulate matter. Similar results were found by Monedero et al. [18] in a parametric study carried out with pine pellets and chips. They observed that excess air reduced gas emissions, but also boiler efficiency, while increasing the secondary air rate reduced CO emissions below the standard limit. In addition, they found the most favorable conditions (with lower gaseous unburned fuels) to be an excess air of 1.4 and a primary/secondary air ratio of 30/70. Afterward, Zadravec et al. [13] found an optimal ratio of primary and secondary air (35/65) that, combined with the elimination of infiltration air and with a low O_2 concentration, leads to lower CO and NO_x emissions and a flue gas sensible heat loss reduction.

Another important factor is the vibration of the grate, which favors the discharge of solid residues into the ashtray and is particularly important for fuels with a tendency to form sintering and deposits such as exhausted olive cake. However, due to periodic disturbances of the fuel bed, vibrating grates can lead to high fly ash and CO emissions as well as incomplete combustion of bottom ash [7] and fluctuations in gas emissions [19]. Silva et al. [20] investigated combustion in a biomass power plant and found that reducing the frequency of grate vibration reduced CO emissions significantly, but also increased agglomeration causing problems in the grate. Therefore, it is essential to find the optimum frequency that favors the removal of residues with minimum emissions.

Finally, also established in the recent ecodesign requirements [10], partial load operation is common in current modular biomass boilers. In general, thermal efficiency is reduced and unburned gas emissions and particulate matter increase when operating at low loads due to inadequate operating conditions [12,21] (which are usually optimized for nominal power). Optimization of some parameters such as excess air, primary/secondary ratio and grate vibration can improve combustion performance and reduce pollutant emissions. González et al. [22] observed that more excess air is required at partial load to assure complete combustion; however, this increases chimney losses and significantly reduces efficiency. In addition, the air/fuel ratio and the primary air flow rate increased as the operating power was reduced, as checked by Lamberg et al. [12], which affects the temperature and residence time of the gases leading to a reduction in the CO oxidation rate. On the other hand, Chandrasekaran [23] reported higher particulate emissions at partial load with wood pellets, which increased when using a fuel with high ash and chlorine content (grass pellets).

Considering the above-mentioned comments, the main objective of the present work is to evaluate the potential of almond shells and exhausted olive cake to be used in domestic boilers by finding out the optimum operating conditions at different loads. An objective function was defined considering CO and PM emissions as the most restrictive variables according to the limits of the regulations.

2. Materials and Methods

2.1. Raw Material and Characterization

Almond shells (AS) and exhausted olive cake pellets (EOC) were used as non-woody biomass. Both wastes come from the food industry; AS comes from the peeling of almonds for consumption and EOC is the solid residue produced when extracting with solvents the remaining oil in olive cake and it is composed of de-oiled peel, pulp and kernels from olive fruit. Table 1 summarizes their physico-chemical characterization. Before the characterization and the combustion experimental tests, particles below 3.15 mm were removed to avoid problems during ignition. Analyses were based on UNE-EN ISO standards for solid biofuels: UNE-EN ISO 18134-2 [24] and UNE-EN ISO 18134-3 [25] for moisture content; UNE-EN ISO 17828 [26] for bulk density; UNE-EN 14780 [27] for sample preparation; UNE-EN ISO 18122 [28] for ash content; UNE-EN ISO 18123 [29] for volatile matter; UNE-EN ISO 16948 [30] for carbon, hydrogen and nitrogen content; UNE-EN ISO 16994 [31] for chlorine and sulfur content and UNE-EN ISO 18125 [32] for heating value.

Table 1. Physico-chemical properties of the fuels tested.

	Almond Shells	**Exhausted Olive Cake**
Moisture (wt. %, wet basis)	13.43	8.39
Bulk density (kg/m^3, wet basis)	398.86 ± 4.99	616.96 ± 0.18
Proximate analysis (wt.%, dry basis)		
Ash	1.93 ± 0.10	8.92 ± 0.41
Volatiles	83.39 ± 4.27	80.85 ± 4.33
Fixed carbon	14.68 ± 4.17	10.16 ± 3.78

Table 1. *Cont.*

	Almond Shells	Exhausted Olive Cake
Ultimate analysis (wt.%, dry basis)		
C	51.53 ± 0.31	50.65 ± 0.20
H	5.18 ± 0.11	5.41 ± 0.10
N	0.72 ± 0.15	1.30 ± 0.10
S	0.01 ± 0.00	0.18 ± 0.01
Cl	0.01 ± 0.01	0.32 ± 0.02
LHV (MJ/kg, dry basis)	18.82 ± 0.02	19.27 ± 0.12

2.2. Experimental Setup and Methodology

2.2.1. Boiler Description

The experimental tests were performed in a three-pass water-tubes, completely monitored fixed bed boiler with a thermal capacity of 55 kW. It is a domestic heating boiler with air staging and automatic ash removal. The full system has been widely described in previous works [18,33]. The feeder system consists of a hopper (previously calibrated for each biomass) where the biomass is loaded, and two synchronized feeding screwdrivers, which allows for changing the biomass flow rate and thus the boiler thermal load. The excess air (λ) is defined as the actual air flow rate with respect to the stoichiometric one. The total air is split into two flows: primary air (up-wards through the grate) and secondary air (through the holes in the wall just above the biomass bed). Two damper actuators operate the corresponding valves to set the desired primary (*PA*)/secondary (*SA*) air ratio. The grate vibration frequency can be manually adjusted between 0 and 45 s for obtaining an adequate ash removal while avoiding bed disturbances leading to fly ash fuel particles.

The fireplace fume temperature (measured just above the combustion chamber, T_{ff}, the closest to location 9 in Figure 1), the chimney fume temperature (T_{cf}, location 11 in Figure 1), and the concentration of O_2, CO, CO_2, NO_x, SO_2, and total hydrocarbons were on-line registered. Gaseous emissions were measured by an Environment medium infrared (MIR) analyzer, which was calibrated before each run with certificated gases mixtures. The particulate matter was iso-kinetically collected by using quartz filters according to UNE-EN 16510-1 [9]. Concentrations and PM were corrected at 10% O_2 according to UNE-EN 303-5 [8].

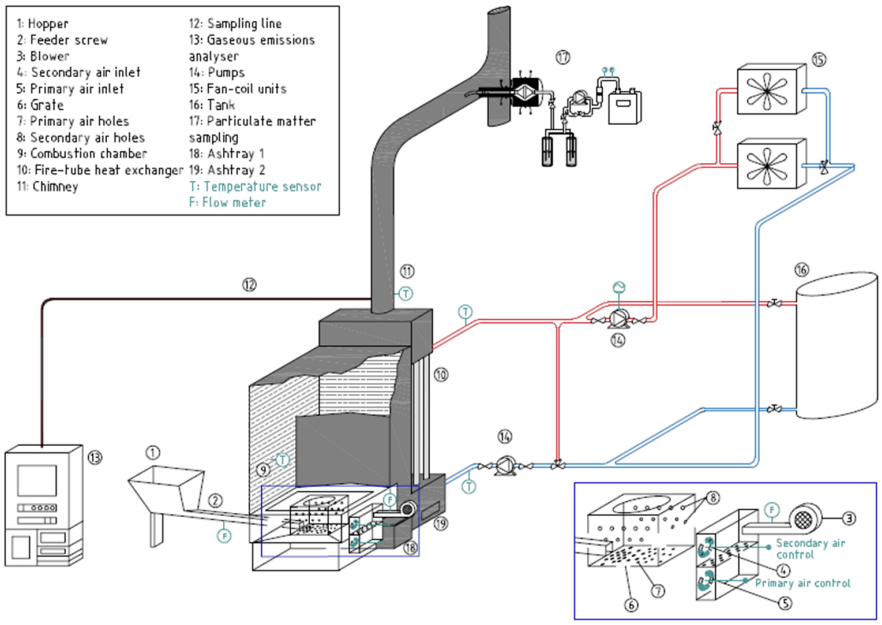


Figure 1. Scheme of the experimental setup.

2.2.2. Methodology

Combustion experimental tests were carried out at two loads (nominal and partial). Nominal load refers to the maximum heat output declared by the manufacturer (55 kW) and partial load is that fulfilling Equation (1) according to UNE-EN 16510-1 [9], thus corresponding to a value of around 24 kW.

$$P_{par} < 0.4 * P_{nom} + 2 \text{ kW} \quad (1)$$

Three boiler operating conditions (factors), considered as the most significant regarding the performance of fixed-bed biomass boilers, were chosen to evaluate the combustion behavior: the excess air (λ), *PA/SA* air ratio, and the grate vibration frequency (V).

Two different types of experimental tests were performed for each biomass and load: an optimization test, and a standard test. The goal of the optimization test was to evaluate the effect of the boiler conditions (excess air, *PA/SA* air ratio and grate vibration) on the combustion behavior by following a methodology based on statistical tools (explained in Section 2.2.3). These experimental tests allowed us to select the optimal operating conditions (leading to lower CO and PM emissions) for each biomass and load. Whereas the purpose of the standard test was to assess and compare the boiler efficiency, solid (PM) and gaseous emissions (CO, NO_x) when fueled with the two types of biomass analyzed.

For each operating condition, once the steady state was achieved and kept for at least 30 min, solid and gaseous emissions measurements were carried out. Steady state was defined fifteen minutes after changes in the flue gas temperature were not higher than 5 °C. The thermal input power, volumetric water flow rate and water temperature gradient (difference between outlet and inlet) were kept constant for each load to make results comparable.

In the case of the standard test, the experimental procedure was slightly different. Once steady state was reached, the operating conditions selected from the optimization tests were maintained for at least 6 h to obtain solid and gaseous emissions and representative fly ash samples for a subsequent study. Gaseous emissions were collected throughout the whole experimental test while particulate matter was collected every 2 h for 30 min periods according to UNE-EN 16510-1 [9].

Once the boiler was cooled down, the solid residue generated at each experimental test was collected. As described in [18,33], the residue was sampled at the following locations (Figure 1): ashtray 1 (below the grate, location 18), ashtray 2 (under the heat exchanger, location 19) and fireplace (the walls and surfaces of the fireplaces, location 9). Representative subsamples of these residues were grounded and analyzed to determine the ash percentage and the unburned matter in the solid residue.

Standard UNE-EN 16510-1 [9] was used to quantify the boiler efficiency (η) that can be calculated from the sensible heat losses in the flue gas (q_a), the latent heat losses in the flue gas (q_b) and the unburned matter in the solid residue (q_c) as shown in Equation (2).

$$\eta = 100 - (q_a + q_b + q_c) \quad (2)$$

The sensible heat loss was estimated from Equations (3) and (4), where $c_{p,fg}$ is the volumetric specific heat of flue gas (kJ/K·m^3), C is the carbon content of the fuel (wt.%), C_r is the carbon content of the residue with respect to the fuel (wt.%), CO and CO_2 are the respective carbon monoxide and carbon dioxide content in dry flue gas (vol.%), $c_{p,fgw}$ is the volumetric heat capacity of water vapor (kJ/K·m^3), H is the fuel hydrogen content (wt.%) and W is the fuel moisture content (wt.%).

$$q_a = \frac{100 \cdot Q_a}{LHV} \quad (3)$$

$$Q_a = \left(T_{cf} - T_a\right)\left(\frac{c_{p,fg} \cdot (C - C_r)}{0.536 \cdot (CO + CO_2)} + c_{p,fgw} \cdot 1.244 \cdot \frac{9H + W}{100}\right) \quad (4)$$

The latent heat loss in the flue gas (q_b) depends on the composition of the latter and was calculated according to Equations (5) and (6).

$$q_b = \frac{100 \cdot Q_b}{LHV} \tag{5}$$

$$Q_b = 12644 \cdot CO \cdot \frac{C - C_r}{0.536 \cdot (CO + CO_2) \cdot 100} \tag{6}$$

Finally, the efficiency loss due to the unburned matter in the solid residue (q_c) was calculated according to Equations (7) and (8), where B is the unburned matter present in the waste with respect to the mass of residue (wt.%) while A is the residue passing through the grate (wt.% with respect to the fuel).

$$q_c = \frac{100 \cdot Q_c}{LHV} \tag{7}$$

$$Q_c = \frac{335 \cdot B \cdot A}{100} \tag{8}$$

2.2.3. Statistical Experimental Design

To find out the optimal boiler operating conditions, a Design of Experiments (DoE) [34] method consisting of two stages was followed: first, a parametric analysis to assess the influence of the operating conditions (factors) and second, an optimization stage, based on the results of the previous stage, seeking to optimize the combustion conditions of the boiler (by decreasing the CO and PM emissions). These stages can be described as follows:

Stage 1: Parametric study: First, the relevant boiler operating conditions (factors) were identified, and a response variable or objective function (*OF*) was defined. The objective function, to be minimized, is shown in equation 9 and it includes both CO and PM emissions, since they are the main drawbacks when working with non-woody biomass (the boiler efficiency not being a challenge). This function represents a quantitative indicator of the combustion behavior, instead of separated analyses of pollutant emissions and efficiency. The objective function was defined considering the limits (denoted as "ref") established in the standard UNE-EN 303-5 for solid fuels for Class 3 boilers (PM_{ref}: 150 mg/Nm^3 and CO_{ref}: 2500 mg/Nm^3). The relative weight of CO was lower than that of PM because the latter is more limiting when dealing with high ash content biomass, as is the case. Therefore, the lowest value of *OF* is desired, with the ultimate goal being below 1, which would mean that the limits are fulfilled.

$$OF = 0.6\left(\frac{PM}{PM_{ref}}\right) + 0.4\left(\frac{CO}{CO_{ref}}\right) \tag{9}$$

The three operating conditions (factors) described in Section 2.2.2 (excess air, *PA/SA* air ratio, and grate vibration) were analyzed. Two levels were defined for each boiler factor, the maximum and minimum allowed by the boiler (λ: 1.2–1.5, *PA/SA*: 20/80–40/60 and V: 20–45 s). The significance of each boiler factor on *OF* was quantified through the p-value (lower than 0.05 show significant statistical differences). The design matrix, coming from a replicated half-factorial experimental design in 3 factors (the three operating conditions), was carried out in the random order shown in Table 2. As observed, the conditions were repeated as suggested by the factorial design for considering the role of repeatability with respect to that of the specific operating parameter.

Table 2. Fractional factorial design (2^3) matrix for the parametric study (stage 1).

Almond Shells and Exhausted Olive Cake								
λ	1.2	1.5	1.2	1.5	1.5	1.5	1.2	1.2
PA/SA	20/80	20/80	40/60	40/60	40/60	20/80	40/60	20/80
V (s)	45	20	20	45	45	20	20	45

Stage 2: Optimization: From stage 1, the less significant boiler operating condition, *V (s)*, was set with the value that showed the best results in stage 1, and the best experimental condition obtained was used as the starting point. Then, a Response Surface Methodology (RSM) [35] was performed. Table 3 shows the design matrix for the optimization study regarding almond shells, again in the random order used (and also showing the replicated tests). Since the *OF* value for exhausted olive cake (shown in the results section) determined from stage 1 was too high (higher than 5), stage 2 was not considered for this fuel. Anyway, and as a reference, the best conditions of stage 1 were considered for its standard test (performance and emissions determination). As is commented in the result section, the high ash content of this type of biomass led to a very inefficient combustion behavior, and further modifications such as technological or compositional (blends with other high-ranking fuels) should be implemented to make this fuel suitable for use in domestic boilers.

Table 3. Design matrix for the optimization study (stage 2).

	Almond Shells						
55 kW	***λ***	1.4	1.5	1.5	1.3	1.3	1.4
	PA/SA	20/80	30/70	10/90	10/90	30/70	20/80
	V (s)	45	45	45	45	45	45
24 kW	***λ***	1.5	1.6	1.6	1.4	1.4	1.5
	PA/SA	40/60	50/50	30/70	30/70	50/50	40/60
	V (s)	20	20	20	20	20	20

3. Results and Discussion

3.1. Statistical Results

- Parametric study (stage 1)

The statistical significance of each boiler factor on the *OF* was determined with the help of the *p*-value. The *p*-values for the two fuels studied and the two loads are shown in Figure 2, along with the critical *p*-value (0.05 in this study). This parameter allows for directly matching changes in the result (i.e., objective function) with a specific operating condition. Values below the critical one indicate statistical significance. Experimental tests for almond shells proved that λ and *PA/SA* were significant under partial load conditions (*p*-values 0.04 and 0.009, respectively). Large values of λ and *PA/SA* air ratio produced smaller values of the *OF* (Figure 3); although, emissions did not fulfil the standard limits ($OF > 1$), and further experimental tests were needed (stage 2). Although none of the operating conditions (factors) were statistically relevant for almond shells under nominal load conditions, the effects of λ and the *PA/SA* air ratio, (with *p*-values of 0.1 and 0.25, respectively) were more significant than that of the grate vibration frequency (*p*-value equal to 0.75). Since the best response for almond shells was observed for λ: 1.5 and *PA/SA*: 40/60 for partial load (~24 kW) and for λ: 1.5 and *PA/SA*: 20/80 for nominal load (~55 kW), these values were selected as starting point for stage 2.

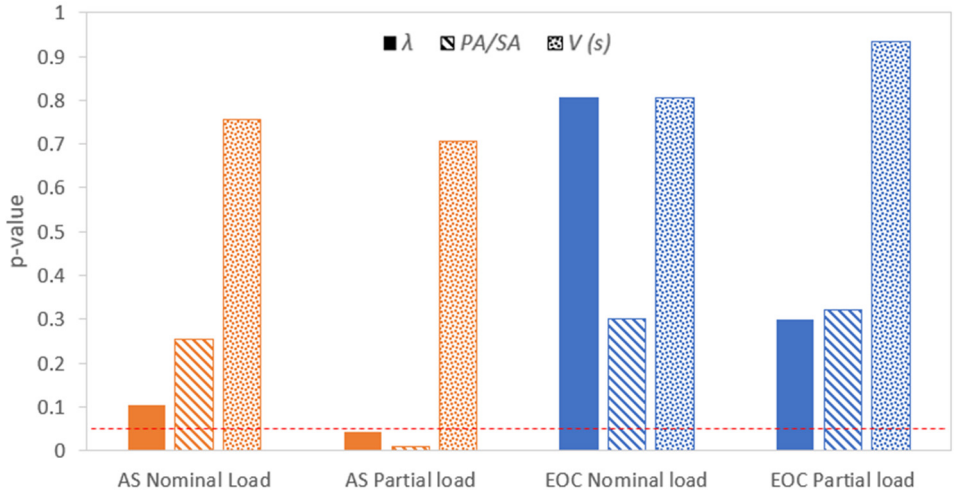


Figure 2. *p*-values for each load and biomass (dashed horizontal line: 0.05 significance level).

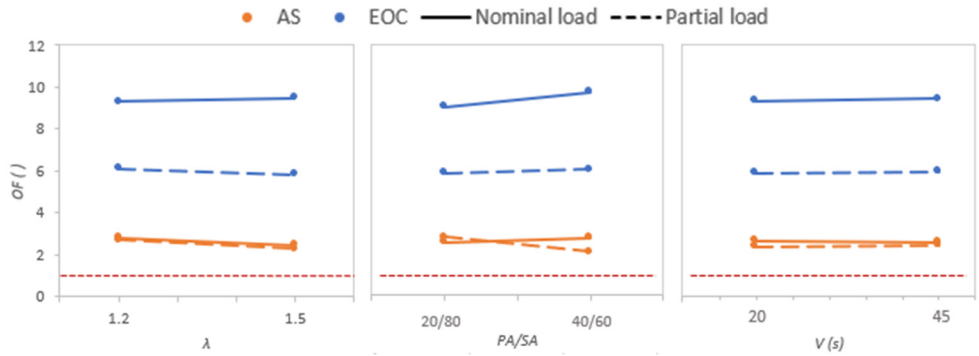


Figure 3. Effect of the factors on the objective function (dashed horizontal line: *OF* value that fulfils the standard limits).

Regarding exhausted olive cake (nominal and partial load), the operating conditions (factors) did not produce significant changes over *OF* (for a significance level of 0.05, Figure 2). Moreover, *OF* values were higher than five for the two loads, meaning that the objective function could not be reached with this fuel (probably because of its extremely high ash content) and, therefore, stage 2 was not performed. Therefore, and as mentioned before, the conditions selected for the EOC standard test for both loads were those with the best *OF* response (excess air 1.5 and a *PA/SA* of 20/80).

To provide additional information, Figure 4 shows the effects of the main significant operating conditions (λ and *PA/SA*) on the efficiency and pollutant emissions for both fuels. Results confirm the expected trends for biomass combustion; an increase in λ improves the combustion behavior by decreasing CO emissions (except for EOC nominal load) and PM emissions (in less magnitude), but also deteriorates the boiler thermal efficiency, because of the higher sensible heat losses [13]. Moreover, NO_x emissions increased (slightly for AS) probably as a result of a better oxidation of fuel nitrogen [33]. Regarding *PA/SA*, the lower *PA/SA* air ratio (20/80), the lower CO and PM emissions for exhausted olive cake. Similar PM emission tendency for almond shells at nominal load was observed while CO emissions were slightly higher at a lower *PA/SA* air ratio. Probably, higher secondary air flow rate improved turbulence and promoted the volatiles oxidation in the secondary combustion zone (Figure 2). This positive effect on reducing emissions was also found by other authors [12,13]. However, both CO and PM emissions at partial load for almond shells were lower when higher a *PA/SA* air ratio was used. The lower air flow at partial load could reduce the air turbulence intensity and then higher primary air was needed for the fuel oxidation. Regarding the *PA/SA* air ratio effect on boiler efficiency, it increased for

exhausted olive cake and it slightly decreased for almond shells when a lower *PA/SA* air ratio was used (in agreement with the CO and PM emission trends commented on above).

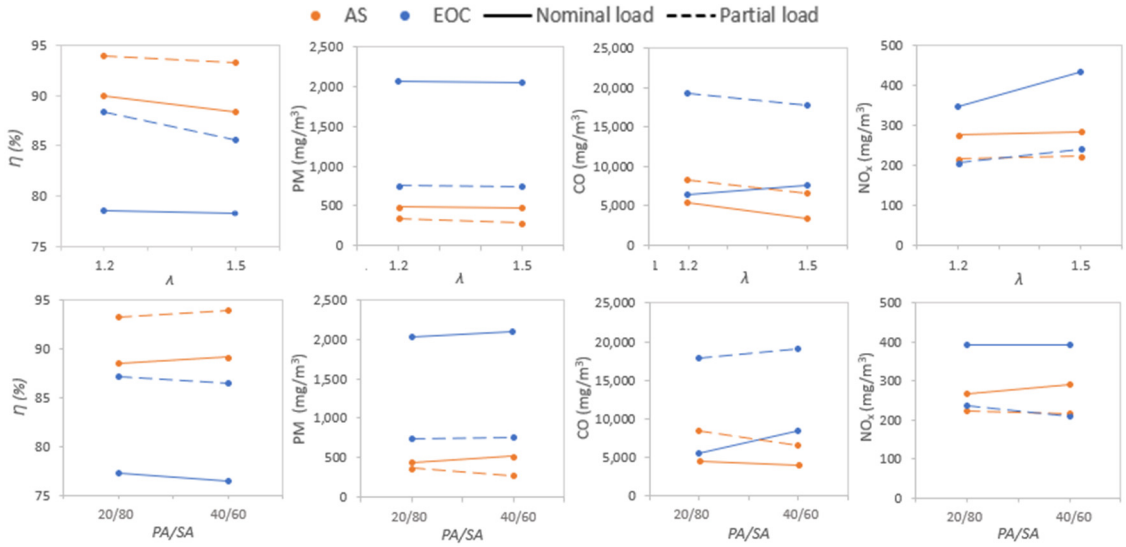


Figure 4. Effects of the excess air (top graphs) and *PA/SA* air ratio (bottom graphs) on the efficiency and pollutant emissions.

- Optimization (stage 2)

As shown in Table 3 and commented on above, since the *OF* for exhausted olive cake was higher than five, stage 2 (optimization) was carried out only for almond shells (nominal and partial load) by performing a factorial design for λ and *PA/SA* air ratio plus two central points at the given value suggested by stage 1, leading to the RSM results shown in Figure 5. For nominal load, λ values were limited by the boiler functionality (the boiler blower did not allow for values greater than 1.5) and, therefore, two λ values up to 1.5 were tested to find the RSM (Table 3).

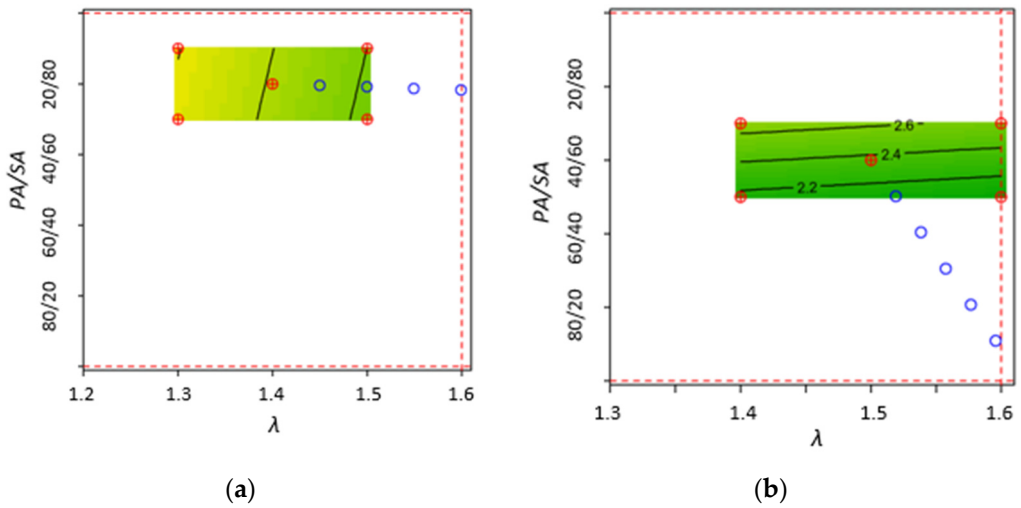


Figure 5. RSM for almond shells (**a**) nominal load, (**b**) partial load.

For nominal load (~55 kW), a first-order model was not significant for the objective function. A quadratic model was also fitted but the adjustment was not improved. Therefore, the operating conditions tested did not allow an $OF \leq 1$. However, the RSM analysis showed a decreasing trend of the *OF* (blue circles). Large values of λ seem to decrease the *OF* while changes in the *PA/SA* air ratio did not show significant changes in the *OF*. Since λ

values greater than 1.5 could not be set in the boiler, a λ of 1.5 and a *PA/SA* of 20/80 were selected as optimum; although, the objective function kept much higher than one in all cases (2.6 for the selected conditions). The optimal excess air (around 1.5) was higher than that typically used for woody biomass (1.4) [18].

Regarding partial load (~24 kW), a statistically significant first-order model was determined. Again, the objective function was higher than one for all the conditions studied and a *OF* decreasing direction was found through the RSM results. The model predicted that the objective function could be reached with higher values of λ (1.6) and the *PA/SA* air ratio (90/10). However, these conditions are far from those tested experimentally and experiments for this operating condition were required. According to this, an experimental test with a λ: 1.6 and the lower secondary air flow allowed by the boiler (50%) was performed and the *OF* obtained (*OF*: 2.0) was similar to that predicted by the model (*OF*: 2.1). Since lower secondary air flow was not allowed by the boiler setting, λ: 1.6 and *PA/SA*: 50/50 were selected as optimal combustion conditions.

Finally, Table 4 shows the selected operating conditions from the statistical study. None of these conditions resulted in emissions (PM and CO) within the limit indicated in UNE-EN 303-5 (for solid fuels). Therefore, the use of this type of biomass as a fuel in domestics boiler is not recommended without any compositional (blended with high-quality biomass) or technological changes (grate, fumes recirculation, etc.). However, almond shell experimental tests (both nominal and partial load) showed an *OF* close to one indicating that this biomass could be probably used blended with small quantities of a high-quality biomass (i.e., wood) to reach the standard limits.

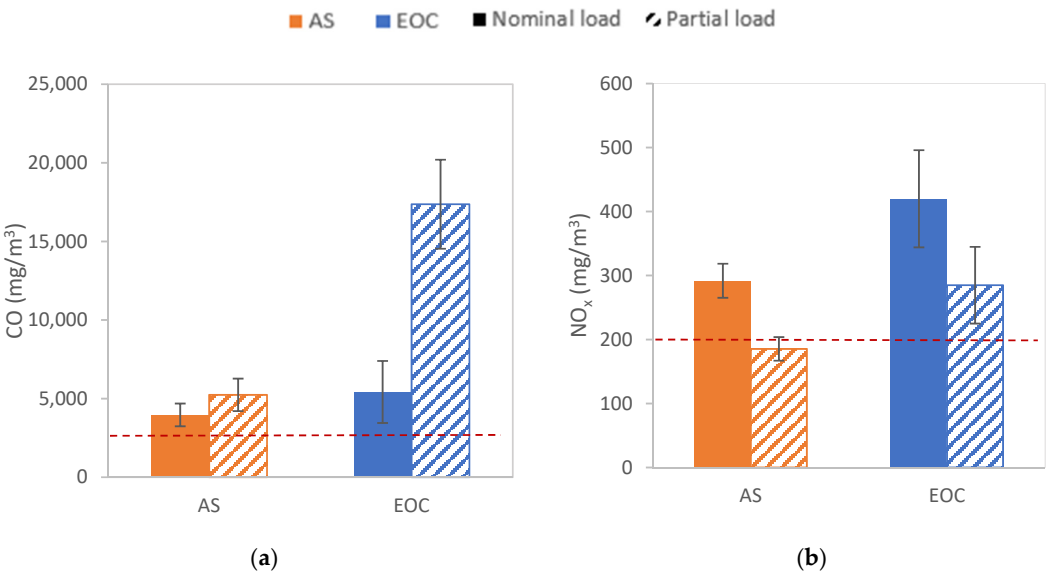
Table 4. Optimal operating condition from the statistical study.

	Almond Shells		**Exhausted Olive Cake**	
	55 kW	**24 kW**	**55 kW**	**24 kW**
λ	1.5	1.6	1.5	1.5
PA/SA	20/80	50/50	20/80	20/80
V (s)	45	20	20	20

3.2. Gaseous and PM Emissions

Emissions were measured under the conditions shown in Table 4 following the standard test described in Section 2.2.2. As is well-known, CO is produced by the partial oxidation of hydrocarbons and, therefore, is generally an indicator of combustion efficiency. Figure 6 shows the CO and NO_x emissions for the fuels and loads tested. CO emissions at partial load were higher than those at nominal load (1.3 times for the almond shells and 3.2 times for the exhausted olive cake). The lower flue gas flow during partial load maybe reduced the turbulence intensity and it deteriorated mixing between the gas coming from the primary combustion zone and secondary air, causing less efficient oxidation of the volatile compounds [36]. The lower fireplace temperatures at partial load (Figure 7), because of incomplete combustion of the fuel, could also contribute to decreasing the oxidation rate of CO [12].

In addition, CO emissions were higher for exhausted olive cake because of its very significant ash content; the considerable accumulated ash in the fireplace may have reduced the diffusion of oxygen through the ash layer towards the carbonaceous residue [33]. It was remarkable to observe the higher T_{ff} (Figure 7) for this biomass, which could have been due to the lower moisture content and also to a possible upward displacement of the combustion zone because of a thicker ash bed (Figure 8b) over the grate (which reduced the primary air supply as mentioned before).



(**a**) (**b**)

Figure 6. (**a**) CO and (**b**) NO_x emissions for each load and biomass (dashed horizontal line: standard limits).

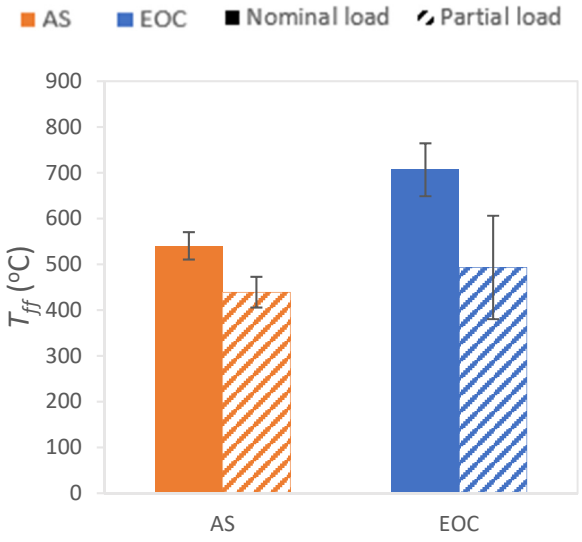


Figure 7. Fireplace fume temperatures for each load and biomass.



(**a**) (**b**)

Figure 8. Images of the fireplace after the experimental test with (**a**) AS, (**b**) EOC at nominal load.

CO results obtained for both fuels and loads were pretty far from those established in the UNE-EN 303-5 [8] limits for solid fuels (2500 mg/m^3 at 10% O_2). Regarding NO_x emissions, these were inversely related to CO emissions as also observed by Van Loo [7]. NO_x emissions during biomass combustion are mainly associated with the oxidation of elemental nitrogen present in the fuel since thermal or prompt NO_x formation is not expected due to the low combustion temperature [37,38]. Instead, NO_x emissions are due to the combustion of the nitrogenous species released with the volatiles and the oxidation of the nitrogen retained in the char.

According to the previous results (higher CO emissions), the less efficient combustion at partial load also led to lower NO_x emissions because of a poorer oxidation of the fuel (Figure 6b) [39]. On the other hand, higher NO_x emissions for exhausted olive cake (Figure 6b) were probably due to its also higher elemental nitrogen (1.3%) [39,40] and the possible catalytic effect of its ash, which also could favor the NO_x formation [41].

Although NO_x emissions are not regulated in UNE-EN 303-5 [8] standard, Regulation (EU) 2015/1189 [10] indicates NO_x emission limits below 200 mg/m^3 at 10% O_2. Thus, only NO_x emissions from almond shells combustion at partial load fulfil the standard. However, because this compound is directly related to the fuel-N, it was not considered in the objective function (Section 2.2.3).

Figure 9 shows PM emissions. For both fuels, lower particulate emissions were observed at partial load, which is probably due to the lower air flow that could reduce the entrainment of fine particles on the flue gas. PM emissions were 2.8 times higher for exhausted olive cake at full load (2.3 times at partial load) when compared to almond shells. This difference could be associated with the higher ash content of the exhausted olive cake. Other authors [23,36,42] also found higher PM emissions for fuels with a high ash content, which were associated with residual fly ash particles ejected from the bed to the flue gas.

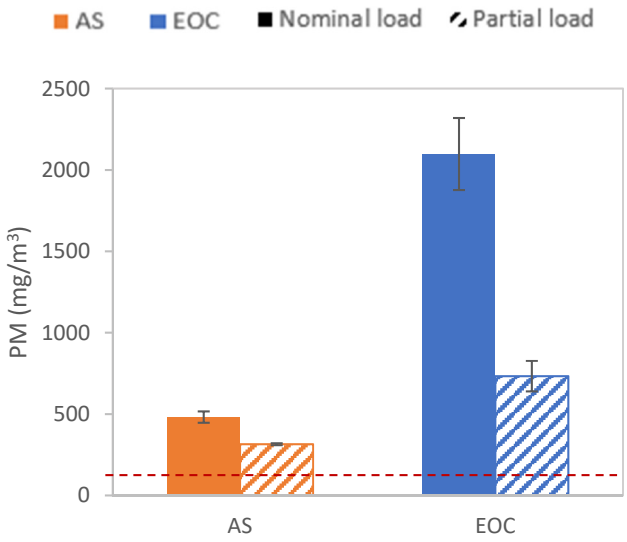


Figure 9. PM emissions for each load and biomass (dashed horizontal line: standard limits).

Moreover, ashes from exhausted olive cake also contain more alkali metals (such as K and Na) as well as chlorine. During combustion, these compounds are volatilized and released into the gas phase [23] contributing to the higher PM emissions of this fuel because of the condensation and nucleation of alkali compounds during gas cooling.

As well as gaseous emissions, the PM emissions obtained for both fuels are outside the limits of UNE-EN 303-5 (150 mg/m^3 at 10% O_2 for wood biomass and 200 mg/m^3 at 10% O_2 for non-woody biomass). However, PM emissions could be reduced by applying some techniques such as the use of a cooling bed or flue gas recirculation [43].

In summary, the gaseous and solid emissions determined for the two non-woody fuels tested do not fulfil the standard UNE-EN 303-5; although, almond shells seem to be

a much more suitable fuel since they could be probably used just blended with a small quantity of a high-quality biomass or additive or by carrying out small modification on the grate. However, exhausted olive cake could not be efficiently used without many further improvements to the boiler design.

3.3. Efficiency

The efficiency loss due to sensible heat in gases (q_a) was higher than that due to latent heat (q_b) and unburned matter in solid residues (q_c) for all fuels and loads except for the case of exhausted olive cake at partial load (Figure 10). For this latter case, a higher (q_b) was obtained due to the significant incomplete combustion as commented in the previous section. For both fuels, the efficiency loss q_a increased at nominal load because of the higher flue gas flow rate and flue gas temperature [21–23]. However, q_b is directly related to the CO emissions, as expected (Figure 6a). Higher q_b but lower q_a was observed for almond shells at partial load, leading to similar efficiency at both loads (around 82%). As commented before, the higher q_b for exhausted olive cake at partial load (even with lower q_a at low load due to the lower T_{cf} achieved) led to a lower efficiency compared with nominal load. Therefore, the efficiency determined for this biomass only fulfils the UNE-EN 303-5 standard at nominal load.

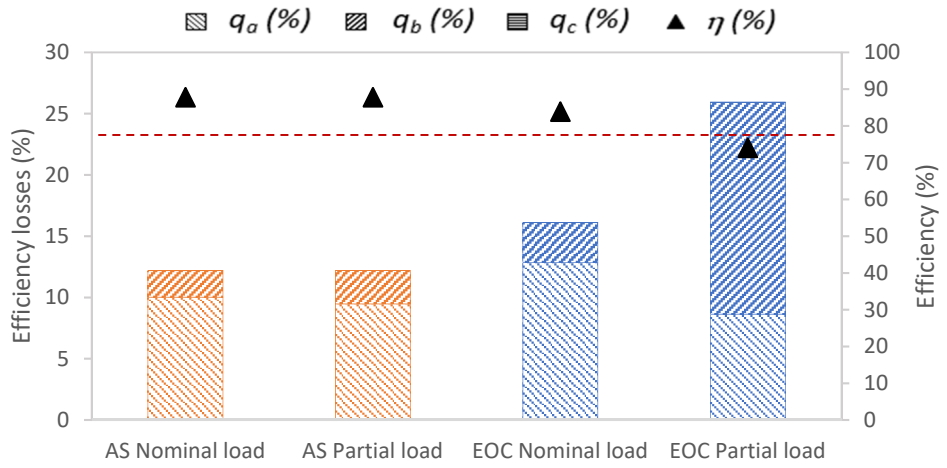


Figure 10. Heat losses and efficiency for each load and biomass (dashed horizontal line: standard limits).

In summary, the boiler efficiency, except for exhausted olive cake combustion at partial load, was clearly above the minimum value of 77%, required in UNE-EN 303-5 for Class 3 boilers [8].

The distribution of solid residues, unburned and ash fractions (%) collected after each experimental test is shown in Figure 11. The residues were mostly formed by ashes (without unburned matter) independently of the fuel tested. As explained, exhausted olive cake ash content (8.9 wt.% d.b) led to a higher amount of residue in the fireplace (Figure 8b), as well as in ashtray 1 and ashtray 2. However, the ash content of residues of exhausted olive cake at nominal load was slightly lower than that of the fuel, which could be associated with the amount of fly ash carried up the chimney (high amount of particulate matter as shown in Figure 9). Regarding the unburned material, the largest amount was collected at partial load with EOC in ashtray 1 (2.8%) and consisted mainly of carbonized pellets that had been reduced in size (without burning) and were able to pass through the grate. A different grate design for this type of fuel could prevent the unburned material from passing through the ashtray. On the other hand, the unburned matter fraction for almond shells formed in the fireplace was below 1%.

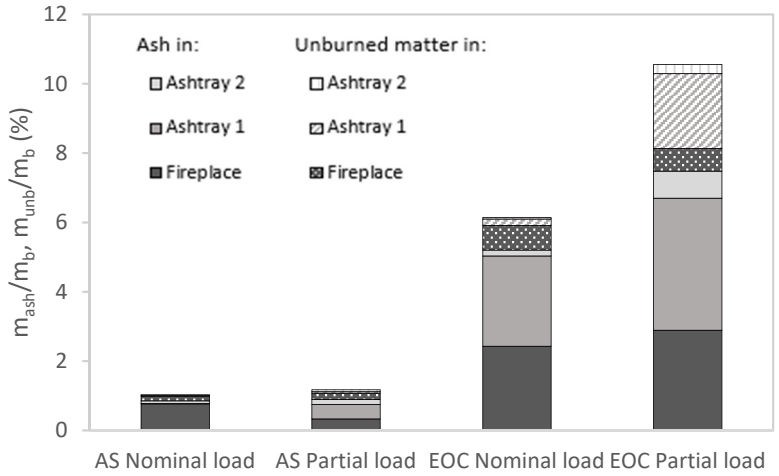


Figure 11. Unburned matter and ash fractions of solid residues collected for the two biomass and loads.

4. Conclusions

This study shows the influence of the operating conditions (excess air, air staging and grate vibration frequency) on the combustion of non-woody biomass in domestic boilers, trying to find out optimal values as well as to provide drawbacks regarding these fuels. Because of their higher sustainability when compared to conventional forest feedstocks, the results could be used by regulators and boilers manufacturers not only for improving boiler design but also to adapt existing standards by considering the characteristics of the new fuels. In fact, existing standards limiting pollutant emissions are mainly based on woody biomass, which restricts the massive use of other more eco-friendly wastes. For this purpose, a statistical analysis based on an RSM was carried out considering CO and PM emissions as the main restrictions when using this type of biomass. The conclusions obtained in this work can be summarized as follows:

- The required excess air (around 1.5) for both loads (~55 and ~24 kW) was higher than that typically used for woody biomass.
- The optimal primary/secondary air ratio at nominal load was 20/80 for both fuels. However, this value changed to 50/50 for almond shells at a low load.
- Because of the very significant ash content of exhausted olive cake, a higher grate vibration frequency was required (20 s with respect to the 45 s needed by almond shells).
- The boiler efficiency at the optimum operating conditions was above the minimum value of 77%, required by UNE-EN 303-5 for Class 3 boilers, except in exhausted olive cake at low load.
- Regarding gaseous and particles emission, CO emissions were lower and NO_x (mainly related to the fuel-N content) were higher at nominal load for both fuels because of a more complete combustion process. Moreover, PM emissions increased due to fuel particles entraining the flue gas.

Despite the acceptable efficiency, the pollutant emissions (CO, NO_x and PM) did not fulfil the UNE-EN 303-5 requirement and, therefore, blending with a higher quality biomass (i.e., pine pellets) and/or using additives to retain particles (such as dolomite, calcite or natural compounds) would be required.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

λ	Excess air
η	Boiler efficiency (%)
LHV	Lower heating value of the fuel (MJ/kg)
T_{ff}	Fireplace fume temperature (°C)
T_{cf}	Chimney fume temperature (°C)
T_a	Ambient temperature (°C)
P_{par}	Partial load power (kW)
P_{nom}	Nominal load power (kW)
q_a	Sensible heat losses in the flue gas (%)
q_b	Latent heat losses in the flue gas (%)
q_c	Heat losses by unburned matter in the solid residue (%)
Q_a	Sensible heat losses in the flue gas (kJ/kg)
Q_b	Latent heat losses in the flue gas (kJ/kg)
Q_c	Heat losses by unburned matter in the solid residue (kJ/kg)
C	Carbon content of the fuel (wt.%)
H	Hydrogen content of the fuel (wt.%)
N	Nitrogen content of the fuel (wt.%)
S	Sulphur content of the fuel (wt.%)
Cl	Chlorine content of the fuel (wt.%)
C_r	Carbon content of the residue with respect to the mass of fuel (wt.%)
W	Moisture content of the fuel (wt.%)
A	Residue passing through the grate (wt.% with respect to the fuel)
B	Unburned matter present in the waste with respect to the mass of residue (wt.%)
m_{ash}	Ash content of solid residues (kg)
m_{unb}	Unburned matter content of solid residues (kg)
m_b	Total fuel feed to the boiler (kg)
AS	Almond shells
EOC	Exhausted olive cake pellet
PA	Primary air flow (%)
SA	Secondary air flow (%)
V	Grate vibration frequency (s)
PM	Particulate matter (mg/m^3)
DoE	Design of Experiments
OF	Objective function
RSM	Response Surface Methodology

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