

Impact of wood combustion in fireplaces on indoor air quality

J. Ascenção¹, S.M. Almeida¹ e V. Martins²

¹ Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, 2695-066 Bobadela-LRS, Portugal. Presenting author email: joao.ascencao@ist.utl.pt

Abstract

The aim of this study was to evaluate the degradation of indoor air quality (IAQ) associated with biomass burning and the exposure of individuals to particulate matter (PM), namely ultrafine particles (PN_{0.01 - 1}), PM₁₀, PM_{2.5} and black carbon (BC), along with other parameters: carbon dioxide (CO₂) and carbon monoxide (CO).

The study was divided into two stages: the first intended to compare the IAQ in rooms with an open fireplace versus a closed fireplace, which included the execution of sixteen experiments, eight of which were carried out in a closed fireplace and the other eight in an open fireplace. The second stage of this study aimed to characterize the IAQ resulting from the combustion emissions of four types of biomass (Eucalyptus globulus, Quercus ilex, Quercus suber and briquettes) in a living room of a house equipped with a closed fireplace.

The results showed that, for both procedures, the limit and guide values, defined by Portuguese legislation and by the World Health Organization (WHO), were exceeded, especially in rooms with open fireplaces, for both gaseous and particulate emissions. For PM it was where the difference was more pronounced, where it was found that, for open fireplaces, the particles of PM_{2.5} were up to 75 times above the legal limit, and for PM₁₀, the values were up to 35 times above the legal limit. For ultrafine particles, BC, CO₂ and CO the values were up between 2 and 9 times higher than those obtained in closed fireplaces. Regarding the second procedure, for all the pollutants, the results revealed peak values for Quercus suber. Eucalyptus globulus and Quercus ilex also stood out punctually, and smaller values were obtained and within the metrics for the burning of briquettes.

Keywords: Indoor air quality, Wood combustion, Fireplaces, PM_{2.5}, PM₁₀

1. Introduction

Although efforts have been made in recent years to reduce air pollution, it still has a significant impact on the population and the environment, with a worrying concentration of various pollutants in urban areas and indoor environments, particularly regarding the impact of the biomass burning in fireplaces.

Especially in colder periods, this heating method is still widely used, and it is an important source of air pollutant, both in outdoor and indoor environments (EPA, 2013), Information on the subject is still very scarce and, knowing that people spend 90% of their time indoors, it makes sense to analyze the impact of biomass burning on the IAQ (Klepeis, 2001). In recent years, there has been an increase in the use of biomass as a heating method, not only to meet domestic demand, but also to promote the use of renewable sources. In the EU, the use of solid biomass for energy production has been strongly encouraged as a measure to mitigate greenhouse gas emissions, replacing fossil fuels. In Portugal, as in many other EU countries, traditional combustion appliances still predominate (Gonçalves, Alves, & Pio, 2012), and in Europe there are more than 70 million outdated fireplaces that, not operate in the most efficient and adequate way, that is, leading to incomplete combustion, having a lower thermal efficiency and a greater environmental impact (Savolahti et al., 2019). This excessive accumulation of indoor pollutants, combined with inefficient ventilation of new building structures, has proved to be highly harmful to human health and the environment. The damage caused by the excessive presence of these pollutants is diverse. The most common effects are usually temporary and consist of discomfort such as eye, nose, skin and throat irritation, acute chest pain, coughing, breathing difficulties, poorer quality of sleep and shorter hours of deep sleep and problems with focus and concentration. These effects, associated with longer exposure, can escalate to more serious conditions, such as asthma, pneumonia, bronchitis and lung and heart problems (Azevedo & Gonçalves, 2011). In extreme cases, atmospheric pollutants can aggravate problems in the neurological, reproductive, and respiratory systems, and may be responsible for the onset of ischemic strokes. Long-term effects are usually chronic, last for years or a lifetime and can, in more extreme cases, lead to death (WHO, 2015).

The impact of pollutants is more significant in the most vulnerable population (the elderly and children), as tissues and organs are in a growing phase in the case of children, or in a more fragile state in the case of elderly people (Mendell & Heath, 2005). Based on this information, we can understand how important is to study these exposure levels and their impact on health. Hence, this report focuses on providing a general overview of the contribution of the main sources of pollutants regarding the degradation of IAQ associated with biomass burning.

2. Methods

2.1. Site description

The sampling campaign was carried out in several places in Portugal as described in the following figure (Figure 1): regarding the first sampling campaign, in Castelo Branco district, 8 sampling tests were carried out, 4 of which took place in houses with an open fireplace and 4 with a closed fireplace. In the district of Lisbon, 7 sampling tests were carried out, 3 of which took place in houses with an open fireplace and 4 with a closed fireplace. In the district of Lisbon, 7 sampling tests were carried out, 3 of which took place in houses with an open fireplace and 4 with a closed fireplace. In the district of Leiria, 1 sampling test was carried out in a house with an open fireplace. Finally, the second



Figure 1. Map of mainland Portugal with indication of the number of residences analyzed per district.

campaign was carried out in Sétubal in a room with closed fireplace and burning of briquettes, holm oak, cork oak and eucalyptus.

2.2. Measurement and sampling conditions

In the first approach, we intended to evaluate and compare the IAQ in residences with an open fireplace and residences with a closed fireplace. For this purpose, sampling campaigns were carried out in 16 homes (eight with an open fireplace and eight with a closed fireplace) where physical-chemical and comfort parameters were measured. The comfort parameters are related to temperature and humidity, and, the physicochemical parameters include ultrafine particles, PM₁, PM_{2.5} and PM₁₀, BC, CO₂ and CO. The biomass used in this procedure varied depending on what was available in the respective house, with arbutus, pine, olive, holm oak and eucalyptus wood being burned. The amount of biomass burned in each sample was approximately seven kilos. The division of the residence where the experiment was carried out was previously ventilated. Additionally, the respective cleaning of the fireplace was previously executed, to be as reliable as possible without any type of contamination. Sampling was carried out with all doors and windows in the room closed during the entire burning process, from the ignition phase to the ash. The time for each sampling ranged from two hours to seven hours.

The second approach aimed to assess the impact of different types of biomass to the IAQ. For this, a study of the concentration of particulate matter was carried out in a room equipped with a closed fireplace. Four different types of biomass were used: briquettes, holm oak, cork oak and eucalyptus. The amount of firewood burned in each sample was approximately seven kilos, identical to the value of the previous procedure. The choice of types of biomass was made based on a statistical analysis of the most used in Portugal, their availability in the market and geographic distribution. The time for each sampling ranged from two to five hours. Sampling was also carried out with all doors and windows in the room closed during the entire burning process. Between changes of biofuels, the ash was removed, and adequate ventil of the room was ensured, applying all cleaning measures before and after each one of the samples.

Both in the first and in the second procedure, all the equipment was placed at a horizontal distance of one meter and twenty centimeters from the fireplace and at a height of half a meter. Furthermore, in both procedures, the ignition phase was carried out in the most ecological and environmental way possible, only with newspapers and a pine cone, in accordance with the recommendations of the Portuguese Environment Agency (EPA, 2014).

2.3. Description of the sampling equipment

In order to measure each pollutant, the following measuring equipment was used: a DustTrak TM DRX Aerosol Monitor 8533 was used to assess particles with an aerodynamic diameter (AD) less than 1 (PM₁), 2.5 (PM_{2.5}), 4 (PM₄) and 10 µm (PM₁₀); a MicroAeth AE51 was used to measure BC concentrations; CO₂, CO, temperature and relative humidity were measured using the TSI IAQ-CALCTM 7545; a TSI Condensation particle model model 3007 was used to measure concentrations of particles with AD between 0.01 and 1 µm;

2.4. Statistical analysis

Statistical tests were performed using the STATISTICA software. Therefore, the Mann-Whitney U test was used, which is a non-parametric test to verify if two samples belong to the same population, in which the different burnings and different types of biomass used were compared. Tests are significant if the confidence interval is p<0.05.

3. Results and discussion

3.1. First sampling campaign - Assessment the impact of open fireplace versus closed fireplace

3.1.1. PM

Figure 2 shows the overall results for PM_{10} for all the rooms equipped with open and closed fireplaces.

For the samples carried out in **open** fireplaces, for **PM**₁₀, all mean concentrations exceeded, to a large extent, the measurements inside the room without burning (marked in red in figure 2). PM₁₀ concentrations were between 14.5 and 7910 μ g/m³, obtaining an average equal to 862.20 ± 579.64 μ g/m³ The concentrations in all samples on open fireplaces, were between five and thirty-five higher, when compared to the values defined by the WHO guidelines and the Ordinance No. 138-G/2021 (50.0 μ g/m³).

Analyzing the samples carried out in **closed** fireplaces, for **PM**₁₀, concentrations were between 7.5 and 788.5 μ g/m³ and the average value was 102.33 ± 74.46 μ g/m³.

It was observed that, in four of the eight measurements, the limit value imposed by the WHO guidelines and Portuguese legislation for PM₁₀ concentrations was exceeded, and the average values of these four measurements were up to five times higher to those limits.





Figure 2. PM₁₀ mass concentrations, in which the red circles represent the mean value of the interior of the room without burning.

As shown in Castro et al. (2018), for PM_{10} , a person who remains in a living room with a burning fireplace will be exposed, on average, to concentrations between 217 to 283 µg/m³, incorporating all phases of ignition and refueling. Furthermore, Canha et al (2018) measured PM_{10} concentrations in open fireplaces and accomplished values between thirteen and twenty-six times higher than the limit imposed by the various competent entities.

Analyzing now **PM**_{2.5} concentrations in **open** fireplaces, the concentrations were between 13.50 and 7845.00 μ g/m³, obtaining an average concentration equal to 850.74 ± 576.16 μ g/m³. These values, obtained in all samples in open fireplaces, were between ten and seventy-nine higher, when compared with the values defined by the guidelines of the WHO and the Ordinance No. 138-G/2021 (25.0 μ g/m³).

In the experiments carried out in closed fireplaces, for PM_{2.5}, the concentrations were between 2.50 and 786.50 μ g/m³, obtaining an average concentration equal to 94.11 ± 76.46 μ g/m³. The concentrations in all samples in closed fireplaces

were between two and ten times higher when compared to the limit imposed by the WHO guidelines and the Ordinance No. 138-G/2021 (25 μ g/m³).

These values were, in general, in agreement with what Canha et al. (2018) determined. Using briquettes and pine as biomass, it was measured for PM_{2.5}, in closed fireplaces, maximum concentrations equal to 86.0 ± 46.3 µg/m³, being approximately three times higher than the limit value $25.0 \,\mu\text{g/m}^3$. As for the experiments in open fireplaces, values between sixteen and nineteen times higher than the legal limit were obtained. Furthermore, in Mcnamara et al. (2017), was carried out an experience in fifty homes in the United States, in rooms equipped with fireplaces, with average PM_{2.5} concentrations of 32.3 \pm 32.6 µg/m³ (ranging from 6.0 to 163 µg/m³). In Canha et al. (2014), it was obtained an average of PM_{2.5} concentrations of 100 \pm 71 µg/m³, being this value far away from the limit value for indoor environments defined by the various national guidelines.

Temporarily analyzing the PM, although the behavior was not exactly homogeneous in all samples, in general, the consumption of biomass showed the existence of several peaks, of which two stand out. It was possible to observe a first peak, existing in practically all the experiments carried out, which corresponds to the primary stage of biomass ignition. In the intermediate stage of combustion, it was where there was a greater discrepancy in values, that can be explained by the various internal and external factors can lead to low quality wood burning. Finally, there was a tendency for a peak in emissions at the end of the combustion, this being associated in particular with the final phase of burning, during which the combustion of a residue that is mainly composed of carbon and ash occurred. These peaks are in line with what was demonstrated in Castro et al. (2018).

3.1.2. Ultrafine particles





Figure 3. Concentrations of ultrafine particles in the sixteen samples carried out, in which the red circles represent the average value of the interior of the room without burning.

PN_{0.01-1} concentrations were significantly higher in experiments performed in open fireplaces (121598 \pm 47793 #/cm³) compared to those performed in closed fireplaces (53592 \pm 32844 #/cm³). The concentrations of PN_{0.01-1}, for open fireplaces, varied between 2516 and 278766 #/cm³. As for the closed ones, values varied between 1476 and 234512 #/cm³, respectively, were obtained.

Both for samplings carried out in open fireplaces and closed fireplaces, the average concentrations of $PN_{0.01-1}$ during the burnings exceeded, to a large extent, the measurements inside the room in the noburning phase (marked in red in figure 3). The concentrations obtained in the measurements performed are in agreement with what was verified in Germany by Salthammer et al. (2014). Furthermore, in the study of Carvalho et al. (2013), there are concentrations of PN, whose maximum values reached 240000 #/cm³. The concentrations recorded were also of the same order of magnitude as those recorded in Press-kristensen et al. (2015), where values around 120000 #/cm³ were obtained. Currently there are no national or international guidelines that allow a more accurate assessment of this parameter. Analyzing the samplings carried out in time, we can conclude that the concentrations in open fireplaces were, on average, twice as high as those obtained in closed fireplaces.

3.1.3. Black carbon





Figure 4. Concentration of BC measured in the sixteen samples taken, in which the red circles represent the average value of the interior of the room without burning.

The percentage of average concentration of BC existing in PM_{10} , in open fireplaces, was 3.12% and, in closed fireplaces, it was 2.90%, being these values being low considering the PM concentration. Both for the samplings carried out in open fireplaces and closed fireplaces, all average concentrations of burning exceeded, to a large extent, the measurements inside the room without burning (marked in red in figure 4).

For the open fireplaces, BC concentrations ranged between 0.17 and 649.63 μ g/m³, with a mean equal to 26.87 ± 9.57 μ g/m³.

For closed fireplaces, it was obtained concentrations between 0.17 and 24.46 µg/m³, with an average of 2.97 ± 1.78 µg/m³. In this study, it was found that, the concentrations of BC in open fireplaces were, on average, nine times higher than those measured in closed fireplaces. The average BC concentrations measured in the fireplaces in the present study were slightly lower than those obtained by Bhaskar et al. (2018), whose records show values between 25 μ g/m³ up to 160 μ g/m³. Also, in Rehman et al. (2011), reports average values of 62 µg/m³, having reached peaks of 1000 µg/m³, meeting the disproportionate peaks obtained in the experiment.

Analyzing the samplings temporally, the highest values of BC concentration were obtained in the initial period of combustion, during the ignition phase. After this initial period, the concentrations, mainly in open fireplaces, suffered a considerable decrease compared to the values initially reached. On the other hand, in closed fireplaces, less linear profiles were observed, with some samples showing a strong oscillation of values during the burning process.

3.1.4. Carbon dioxide





Figure 5. CO₂ concentration recorded in the sixteen samples taken, in which the red circles represent the mean value of the interior of the room without burning.

In open fireplaces, CO_2 concentrations varied between 345 and 2176 ppm, with an average equal to 594.58 ± 169.22 ppm (1065.00 ± 365.07 mg/m³). The average concentrations of CO_2 , resulting from open fire burning, were generally in line with the measurements inside the room in the no-burning phase (marked in red in Figure 5), where only samples 4 and 8 stand out.

For closed fireplaces, concentrations were ranging between 400.50 and 1379.50 ppm were obtained, with an average of 742.35 ± 249.14 ppm (1345.87 ± 249.14 mg/m³). It was observed that, although the highest value was reached in an open fireplace, the average values were higher in closed fireplaces, being, on average, 1.25 times higher than those measured in open fireplaces. The results of a higher concentration of CO₂ in closed fireplaces would be expected given that we have better conditions to achieve complete combustion, in which the products of the same will be CO₂ and H₂O. These concentrations are in line with the values determined by Castro et al. (2018), which reports having recorded average CO_2 values of 480 ± 110 ppm. Canha et al. (2018) recorded average values of 947 \pm 170 mg/m³ for the burning of pine and 1040 \pm 140 mg/m³ for the burning of briquettes.

By observing the temporal evolution of CO_2 concentrations, we can conclude that exists a tendency of the increase of CO_2 in the initial combustion phase, during which, in almost all of them, the maximum concentration value was reached. In CO_2 , we can find a more heterogeneous

profile, in which some experiments showed an everincreasing profile of increased concentration, and others the reduction and stabilization of emissions.

3.1.5. Carbon monoxide





Figure 6. CO concentration recorded in the sixteen samples taken, in which the red circles represent the mean value of the interior of the room without burning.

In open fireplaces, CO concentrations varied between 1.00 and 30.05 ppm, with an average equal to 5.54 ± 3.18 ppm (6.29 ± 3.60 mg/m³). For closed fireplaces, concentrations ranging between 2.34 and 1.05 ppm were obtained, with an average of $2.34 \pm$ 1.05 ppm (2.69 ± 1.19 mg/m³). For CO, it was possible to conclude that the highest value was registered in an open fireplace, and, on average, the values were twice as high as those presented in closed fireplaces. Either in the samplings carried out in open fireplaces and in those carried out in closed fireplaces, all the average concentrations of burning exceeded those measured inside the room without burning (marked in red in figure 6). These values are in agreement with what was determined in Canha et al. (2018), with average values for burning pine of $6.02 \pm 3.33 \text{ mg/m}^3$ and $5.55 \pm 4.72 \text{ mg/m}^3$ for burning briquettes in open fireplace. Also in Salthammer et al. (2014), the average CO values are in line with those presented in this study, with an average concentration, within half an hour, of around 5.11 mg/m³ for open fireplaces and 3.24 mg /m³ for closed ones. In Chowdhury et al. (2013), it was obtainded values ranging between 3 and 11 ppm. Temporally, it was possible to observe that there was a wide variety of peaks throughout the procedure, being classified as a very diverse and heterogeneous behavior.

3.1.6. Temperature and relative humidity

Starting with the temperature, in open fireplaces, the temperature ranged between 13.75 and 46.00 °C, with an average equal to 26.14 ± 3.42 °C. As for the closed fireplaces, temperatures were ranging between 14.45 and 38.40 °C, with an average of 22.58 ± 1.69 °C. It was possible to observe that there was no great discrepancy in the mean values achieved, and, the highest value was recorded in an open fireplace, at the height of flame combustion. The minimum values were always recorded in a very embryonic stage of burning, namely in the ignition stage. Temporally, both in open and closed fireplaces, the temperature behavior was very homogeneous, with a rapid rise in the initial stage of combustion. There was still a considerable permanence at high values in the flame combustion stage, later occurring a decrease when we enter the final stage of flameless combustion.

Regarding the relative humidity, we can report that, for open fireplaces, we obtained values ranging between 11.35 and 74.00 %rh, with an average equal to 38.67 ± 12.03 %rh. As for the closed fireplaces, we obtained values ranging between 17.06 and 69.60 %rh, with an average of 48.15 ± 7.47 %rh.

We can assume that, in general, there was the presence of higher average values in closed fireplaces. Nonetheless, both the highest and the lowest relative humidity values were achieved in a sample from an open fireplace. The highest values were registered in the ignition phase and a substantial drop occurred when we entered the flame combustion phase. As the burning process came to an end and the transition to the final stage began, there was an increase close to the values recorded initially.

3.2. Second sampling campaign - Assessment of the impacts of different types of biomass







Analyzing the PM, it was possible to conclude that the large concentration of particles was found represented in the fine fraction. Regarding the PM_{2.5} concentrations, the highest were registered in the cork oak burning and the lowest were recorded in the eucalyptus burning. We can conclude that in all burnings the legal limit imposed by the WHO guidelines and the Ordinance No. 138-G/2021 (25.0 $\mu g/m^3$) was exceeded. In the case of the cork oak, values were recorded about eleven times higher $(248.03 \pm 118.13 \mu g/m^3)$. As for briquettes and eucalyptus, were registered average values of 42.26 \pm 8.23 µg/m³ and 47.79 \pm 5.03 µg/m³ were recorded, respectively. For holm oak, the values were a little higher and are also in agreement with what was recorded in some samplings of the first campaign, obtaining an average concentration of PM2.5 of 72.20 \pm 37.11 µg/m³. For PM₁₀, the limit values were also exceeded in all burns, following the same trend recorded in PM2.5. The highest value was obtained in the burning of cork oak (270.41 \pm 119.75 μ g/m³), being a value approximately five times higher than the legal limit imposed by the WHO guidelines and

Ordinance No. 353-A/2013 (50.0 μ g/m³). For eucalyptus and briquettes, concentrations were very close to the legal limit, with mean values of 54.57 ± 8.37 μ g/m³ and 57.31 ± 13.10 μ g/m³ respectively. For holm oak, a mean concentration of 88.60 ± 41.24 μ g/m³ was recorded.

3.2.2. Ultrafine particles



Figure 8. PN_{0.01-1} concentration in the different samplings performed.

The concentrations of PN_{0.01-1} were higher in cork oak burning, for which an average value of 49012.50 \pm 23639.26 #/cm³ was obtained. The lowest value was obtained in the burning of holm oak, with an average concentration of 26433.61 \pm 5918.33 #/cm³. As for the briquettes and eucalyptus, there were mean concentrations of 45540.05 \pm 25979.94 #/cm³ and 38383.56 \pm 19013.21 #/cm³ respectively. It was also concluded that the values obtained in the different burns were between three and six times higher when compared to the reference value (average concentration of particles inside the room without burning).



Figure 7. BC concentration in the different samplings performed. In BC, there were again higher concentrations in cork oak burning, with an average value of $3.76 \pm 1.53 \ \mu\text{g/m}^3$ being obtained. The lowest value was obtained in the burning of briquettes, with an average concentration of $1.01 \pm 0.83 \ \mu\text{g/m}^3$. For holm oak and eucalyptus, mean concentrations of $1.41 \pm 1.01 \ \mu\text{g/m}^3$ and $1.49 \pm 0.14 \ \mu\text{g/m}^3$ respectively were recorded. We can also conclude that the values obtained in the different burns were between one and five times higher when compared to the reference value (average BC concentration inside the room without burning).





Figure 10. CO₂ concentration in the different samplings carried out and representation of legal limits.

Analyzing now the CO₂, we can see that the highest values were registered in the burning of the cork oak, in which an average value of 1360.05 ± 245.54 ppm was obtained. The lowest value was obtained from burning eucalyptus, with an average concentration of 980.07 ± 191.22 ppm. As for holm oak and briquettes, mean concentrations of 1254.62 ± 207.50 ppm and 1074.66 ± 234.47 ppm were recorded, respectively. We can observe that, only in the burning of cork oak, the legal limits were exceeded: either the limit imposed by Ordinance No. 138-G/2021 (1250 ppm) or the limit imposed by the WHO (1000 ppm). Comparing the results obtained with the WHO guidelines, only eucalyptus burning did not reach this maximum, despite being very close to it.

3.2.5. Carbon Monoxide

For CO, we can observe that the highest values were registered in the burning of the cork oak, in which an average value of 1.27 ± 0.43 ppm was



Figure 11. Concentration of CO in different samples.

obtained. The lowest value was obtained in the holm oak burning, with an average concentration of $0.51 \pm$ 0.19 ppm, being even below the reference value (average concentration of CO inside the room without burning occurrence). As for eucalyptus and briquettes, average concentrations of 0.71 ± 0.28 ppm and 0.58 ± 0.23 ppm were recorded, respectively. We can observe that, in any burning, the legal limits imposed by Ordinance No. 138-G/2021 were exceeded (9 ppm for a period of 8 hours and 6 ppm for 24 hours).

3.2.6. Comfort parameters: Temperature and Relative Humidity

Finally, analyzing the comfort parameters, we can observe that, for temperature, the highest values were registered in the burning of briquettes (figure 34), during which an average value of 23.85 ± 1.70 °C was obtained. The lowest value was obtained in the cork oak burning, reaching an average value of 21.36 ± 0.64 °C. For eucalyptus and holm oak, mean values of 22.58 ± 2.35 °C and 21.77 ± 0.33 °C, respectively. For relative humidity, we had the highest value being registered in the burning of cork oak, in which an average value of 62.88 ± 1.74 %rh was obtained. The lowest values were registered in the burning of briquettes and eucalyptus, with an average value of 57.25 ± 2.70 %rh and 57.34 ± 3.66 %rh, respectively. As for the holm oak, an average value of 60.31 ± 1.93 %rh was obtained.

4. Conclusion

The results obtained in this study allowed us to conclude that the biomass combustion process in indoor environments is a highly polluting and complex process. It was also found that pollutant emissions in open fireplaces were much higher than those measured in closed fireplaces. The various guidelines imposed, either by Portuguese legislation or by the WHO, were exceeded, especially in open fireplaces, not only in gaseous emissions, but, with particular emphasis, in particulate matter.

Therefore, a wide range of values was found throughout the different phases of the burning process, with a still considerable diversity of behavior in the time series, whose explanation can be found in the countless operational parameters and external elements mentioned above. It was possible to observe, for example, in some burnings, that the ignition phase had emissions with an order of magnitude higher than those observed during the flame phase. In some burnings, concentration peaks were also observed during the final burning phase, where the burning of carbon residue occurs, these values being higher than those obtained in the flame phase, which emphasizes the heterogeneity of behavior described and the complexity of the process of burning.

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