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Impact of Biomass Home Heating, Cooking Styles, and Bread Toasting on the Indoor Air Quality at Portuguese Dwellings: A Case Study

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Abstract: This study evaluated the emissions of specific indoor sources usually present in Portuguese dwellings in order to understand their impact on the indoor air quality. With this aim, three typical activities were studied including home heating using two types of fireplaces (open and closed) and biofuels (pinewood and briquettes), cooking styles (frying and boiling) in different types of kitchen appliances, and several levels of bread toasting. The levels of specific pollutants were found to be above the established Portuguese limit values including VOCs, formaldehyde, and particulate matter ($PM_{2.5}$ and PM_{10}). Although these emissions are transient and short in duration, the resulting concentrations are high and can severely impact the occupants' daily exposure. Besides promoting good ventilation, the choice of residential appliances with low emissions should be taken into account. In addition, it is important that occupants perform specific activities following the best practices so that their exposure to pollutants is minimized.

Keywords: indoor sources; dwellings; indoor air quality; wood combustion; cooking emissions; particulate matter

1. Introduction

The impact of indoor air quality (IAQ) on occupants' health and welfare is a subject that gathered the attention of the scientific community over the last two decades [1]. There has been extensive research on micro-environments where people spend their time with the aim of understanding exposure patterns. Initially, the studied micro-environments were mainly buildings in which people spend a significant fraction of the daytime such as offices [2,3], public buildings like schools [4–6], dwellings [7], and other indoor places where leisure activities [8–10] are developed. In recent years, the need to understand the exposure levels in an integrated perspective has started to gain consensus since individuals spend around 90% of their time in different indoor environments. Therefore, considering only one micro-environment is not representative of the daily overall exposure. Understanding the individuals' exposure during the different periods of the day in specific micro-environments is fundamental for providing a real and an integrated assessment [11–13].

Residential indoor spaces were found to be micro-environments with the highest contribution to the daily exposure to particulate matter. A study in Korea, where the contribution of several micro-environments for the personal exposure was assessed, showed that residential indoor locations contributed to the daily individual $PM_{2.5}$ exposure with a total of 55.4% in the summer and a total of 49.5% in the winter while PM_{10} exposure contributed a total of 50.0% in the summer and 43.0% in the winter [14].

Several studies have focused on different indoor sources at residential micro-environments that can increase the individuals' exposure. These indoor sources may be associated with specific frequent activities carried out by dwellers, such as cooking [15,16], cleaning [17], candle burning [18], home heating [19–22], vacuuming [15], and other activities, which increase pollutants above acceptable indoor levels. A wide range of pollutants can originate via these activities including particulate matter ($PM_{2.5}$, PM_{10}) [23], ultrafine particles (UFP) [15,24], volatile organic compounds (VOCs) [25,26], carbon dioxide [19], carbon monoxide [19,22], formaldehyde [19], polycyclic aromatic hydrocarbons [16,27], among others. For instance, cooking was found to increase PM levels by four times the background levels in a study at 14 naturally ventilated homes of Hong Kong [28]. A review on which factors impact emissions/concentrations of cooking generated particulate matter from controlled studies found that the cooking method, type and quality of energy (heating) source, burner size, cooking pan, cooking oil, food, additives, source surface area, cooking temperature, ventilation, and position of the cooking pan on the stove are influential factors affecting PM emission rates and resulting concentrations [23]. Typical household appliances such as flat irons and hair dryers have shown to promote significant emissions of particles in test chambers with mean diameters below 100 nm where maximum emissions of UFP could be found only after a few minutes of the appliances being switched on [24]. Other pollutant emissions found in these appliances were VOCs. Among these, cyclic siloxanes dominate the emissions from flat irons.

To perform integrated exposure assessments, it is important to gather information regarding the contribution of specific human activities to IAQ. This research aims to give insight into this issue by providing a case study of different household activities and their impact on IAQ and occupants' exposure levels during the worst ventilation conditions, i.e., the worst-case scenario. The selected activities were different cooking styles and house heating with biofuels.

2. Experiments

2.1. Selection of the Studied Conditions

In order to understand the main types of dwellings and which type of activities were performed by their occupants, an online questionnaire was conducted over three months with a total of 296 individual answers from 17 of the 20 Portuguese districts. The two districts that gathered more answers were Setúbal (43%) and Lisbon (38%).

Two types of dwellings were found to be predominant among all participants, which include apartments (61%) and detached houses (39%). This fact is confirmed by the national housing inquiry done by the Portuguese Institute of Statistics, which pointed out these two types of dwellings as prevalent in Portugal [29].

The questionnaire carried out in the framework of this study also tried to understand some behaviors/habits of the participants. Some results from the study are below.

(1) 57% of the participants sleep with the bedroom door closed;
(2) 8% of the participants stated that someone smokes indoors;
(3) 92% use the toaster daily and 60% use scented products at home;
(4) 63% stated that they spend between 8 h and 13 h at home (with only 16% stating that spend less than 8 h at home);
(5) 76% usually open the windows between six to seven times per week;
(6) 71% of owners clean the house once a week while the remaining do it more often;
(7) Grilled (38%), fried (35%), boiled (13%), stewed (8%), and roasted (5%) meals represent the most frequent cooking styles;

(8) The most commonly used energy sources to cook are natural gas (37%), bottled propane gas (23%), electricity (19%), and others (22%).
(9) The four main heating systems in detached houses are open fireplace (37%), which is followed by air conditioning (29%), fireplace insert (28%), and oil heater (26%) while, in apartments, the main heating systems are oil heaters (28%), air conditioning (17%), fireplace insert (12%), and open fireplaces (10%).

Based on these results, it was decided to focus our study on detached houses and apartments and to study activities such as house heating (using open fireplace and fireplace insert) and frequent cooking methods (fried and boiled cooking and toasting). Grilling was not chosen since, usually, it is done outdoors. These heating systems and cooking set-ups represent the ones used by the Portuguese population. Given the similarities of both climate conditions and life styles in Southern European countries, it may be considered that these activities/sites are also representative of the entire Mediterranean region.

2.2. Study Site

The studied dwellings were located in an urban and semi-urban area (parish of Pinhal Novo) of Setúbal district, Portugal. Setúbal is the third most populated district of the country with a total of 849,842 inhabitants, which represents around 8.1% of the Portuguese population [30]. Table 1 presents the characteristics of the three studied dwellings. Figure 1 depicts the floor plans and highlights the rooms (kitchen and living room) where measurements took place. In the inhabited dwellings, none of the occupants were smokers.

Table 1. Characteristics of the studied dwellings.

Characteristic	Dwelling 1	Dwelling 2	Dwelling 3
Type	Apartment—two bedrooms	Detached house with four bedrooms	Terraced house with four bedrooms
Floor	Third	Ground	Ground and first
Area (m^2)	110	166 (within a private land of 5500 m^2)	180 with a small backyard
Height (m)	2.80	2.80	2.80
Construction year	1999	1983	1990
Walls	Brick with thermal isolation (air box)	Brick without any isolation	Brick without any isolation
Windows	Single-hung aluminum windows	Single-hung wood windows	Single-hung aluminum windows
Floor	Tiles (living room, hallway and kitchen) and parquet wood (bedrooms)	Tiles	Tiles
Gas	Piped natural gas	Bottled butane	Piped natural gas
Ventilation	Natural	Natural	Natural
Surroundings	Urban area, located near a moderate traffic street and with a restaurant on the ground floor	Semi-urban area, located near a low traffic street	Semi-urban area, located near a low traffic street
Use	Inhabited by 2 persons	Inhabited by 2 persons	Uninhabited and without furniture

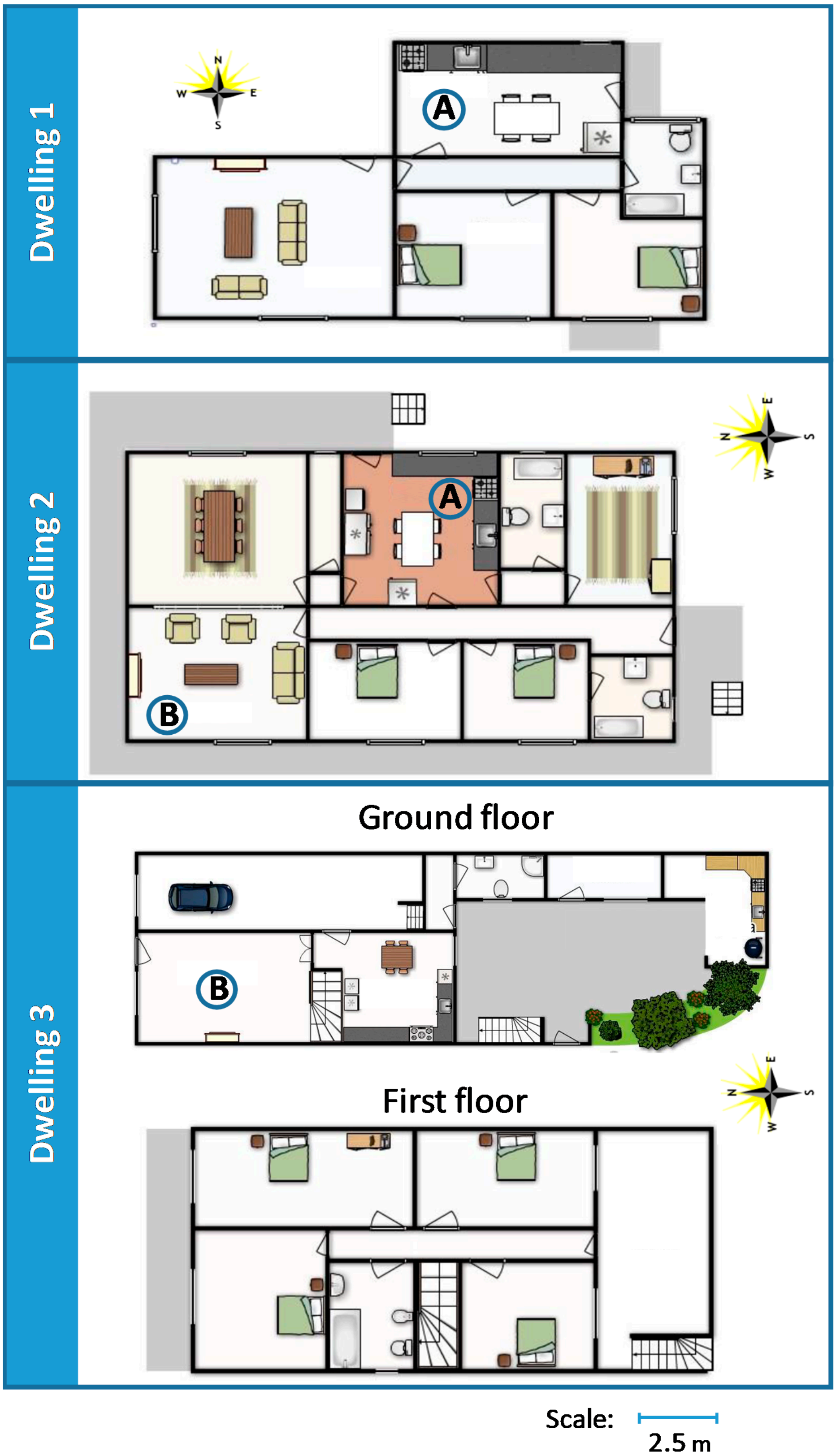

Figure 1. Floor plans of the studied dwellings with specification of the rooms where the monitoring was conducted: A—kitchen and B—living room.

Characterization of the Monitored Rooms

In the houses described above, the kitchens and living rooms were monitored. Table 2 presents their characteristics.

Table 2. Characteristics of the studied rooms.

Room	Dwelling	Characteristics
Kitchen	Dwelling 1 	- 1 window, one inner door to hallway, and one balcony door - Area: 18 m^2 - Volume: 50 m^3 - Fully equipped with appliances
	Dwelling 2 	- 1 window, three inner doors to hallway, and one door to the courtyard - Area: 24 m^2 - Volume: 67 m^3 - Fully equipped with appliances
Living room	Dwelling 2 	- One window and one door to hallway - Area: 30 m^2 - Volume: 84 m^3 - Open fireplace - No rugs or carpets - Furniture and electronic devices
	Dwelling 3 	- One window and two doors (one to the street and another to the kitchen) - Area: 30 m^2 - Volume: 84 m^3 - Fireplace insert - No rugs or carpets

It is worth mentioning that, despite that a total of three dwellings were considered in this study, each activity was investigated in only two houses. The cooking styles and toasting were evaluated in two kitchens from two different types of dwellings including an apartment (dwelling 1) and a detached house (dwelling 2). However, for the study of biomass burning emissions, it was not possible to find an apartment with fireplace (open fireplace or fireplace insert). From our preliminary inquiry of 296 participants, only 10% to 12% of the apartments had fireplace inserts or open fireplaces, respectively.

Therefore, the same type of dwelling (detached houses, dwellings 2, and dwellings 3) was chosen to perform the experiments for which the main focus was to study different types of biofuels and combustion devices.

2.3. Domestic Activities

2.3.1. Biomass Burning Emissions

The monitoring of biomass burning emissions was conducted using two types of appliances—an open fireplace and a fireplace insert—in living rooms of dwellings 2 and 3, respectively (see Figure 2).

Figure 2. Appliances used for biomass burning include (**left**) an open fireplace located in the living room of dwelling 2 and (**right**) a closed fireplace (insert) located in the living room of dwelling 3.

In each device, two biofuel types were combusted using pinewood and briquettes, which were both bought at the local market. According to the producer's specifications, briquettes were 100% composed of pressed wood with a calorific value of 4800 kcal/kg, 5–7% humidity, and 3% ashes. Each test was conducted on a daily basis with the opening of windows and doors for two hours prior to the experiment in order to promote renovation of the living room indoor air. During the experiments, all doors and windows were closed. A total of 5 kg of each biofuel was burned in one go per experiment. Ashes were removed before the following combustion cycle. For each biofuel and appliance, the experiments were repeated on three different days in June 2015. Each combustion cycle lasted between 1 h and 1 h and a half. A total of 12 experiments were conducted. Experiment 2 of the combustion conditions CP (pinewood in a closed fireplace) was discarded due to ignition problems.

Experiment OP (pinewood in an open fireplace) was conducted from 8 July 2015 to 10 July 2015. Experiment OB (briquettes in an open fireplace) was conducted from 11 to 13 July 2015 while experiment CP (pinewood in a closed fireplace) was conducted from 18 to 20 July 2015. Experiment CB (briquettes in a closed fireplace) was conducted from 21 to 23 July 2015. All experiments were conducted between 19:00 and 23:00, local time.

2.3.2. Cooking Emissions

Two different ways of cooking, which are frying and boiling, were carried out in three different appliances including the electric cooking plate, the gas cooking plate (using bottled butane), and the gas stove (with piped natural gas). Figure 3 presents the type of meals prepared and the cooking utensils and appliances used. Each meal was prepared three times on different days for more than 30 min. The kitchen exhausts were not connected during the experiments. A total of 24 experiments were conducted in July 2015.

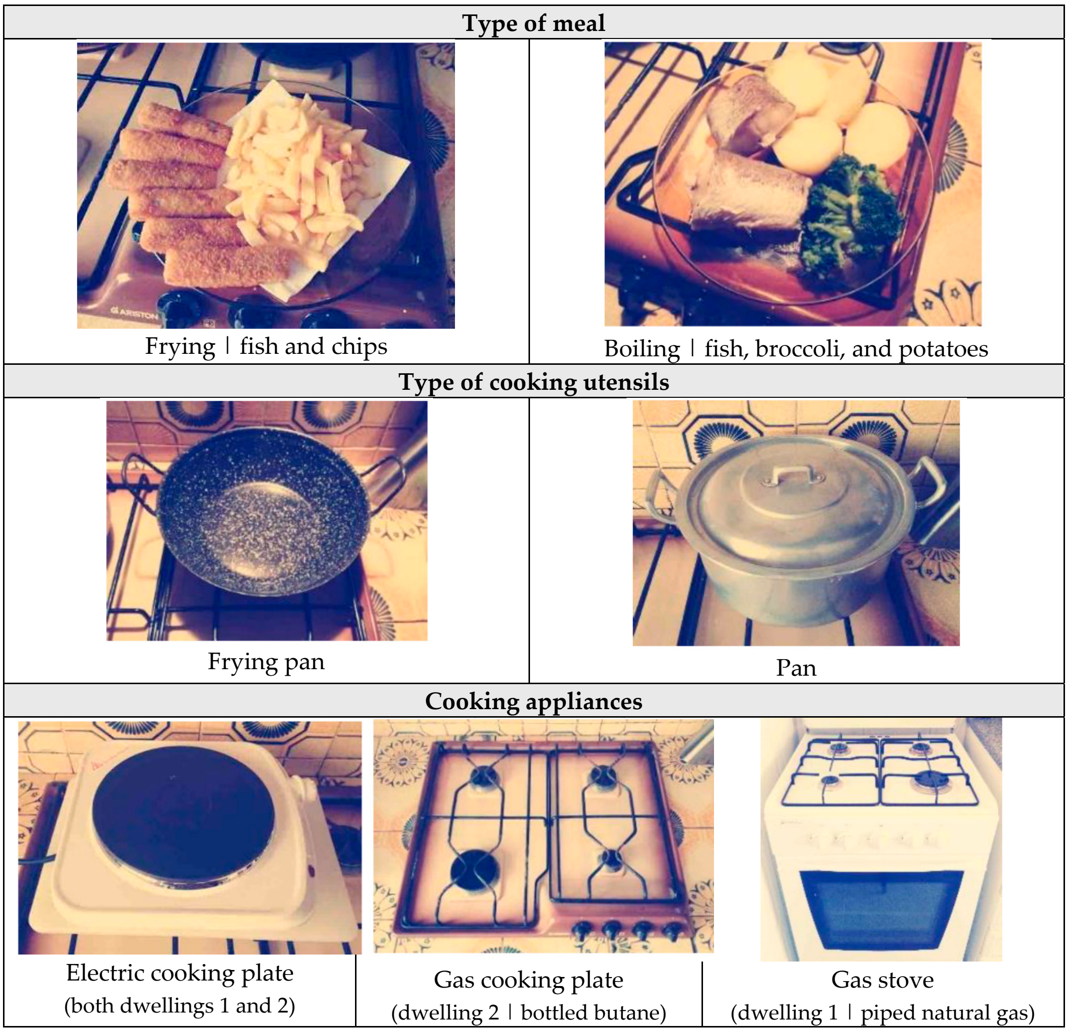

Figure 3. Type of meals prepared and cooking utilities and appliances used.

2.3.3. Emissions from Toasting

To evaluate three different degrees of toasting (slight, medium, and heavy, which is shown in Figure 4), a Philips toaster was used. The experiments were conducted in dwellings 1 and 2 in June and July 2015. For each level of toasting, a total of five experiments were performed for periods of 30 min over three days in each dwelling.

Figure 4. Levels of loaf bread toasting studied (**right**: slightly, medium, and heavily toasted) using a Philips toaster (**left**).

2.4. Indoor Air Quality Monitoring

IAQ monitoring was performed using real time instruments.

- Graywolf (IQ-610 probe, WolfSense Solutions, Shelton, CT, USA) to measure CO_2 (measuring range: 0 to 5000 ppm, accuracy: ±3% of reading +50 ppm), total volatile organic compounds—VOCs (measuring range: 5 to 20,000 ppb, resolution of 1 ppb), CO (measuring range: 0 to 500 ppm, accuracy: ±2 ppm < 50 ppm, ±3% reading > 50 ppm), temperature (measuring range: −25 °C to 70 °C, accuracy: ±0.3 °C) and relative humidity—RH (range: 0 to 100%, accuracy: ±2% < 80 %, ±3% > 80%);
- Formaldemeter (htV-M, PPM Technology, Wales, UK) to measure formaldehyde—CH_2O (measuring range: 0–10 ppm, accuracy: 10% at 2 ppm);
- DustTrak monitor (8530 model, TSI, Shoreview, MN, USA) to measure PM_{10}, $PM_{2.5}$, and PM_1 (measuring range: 0.001 $mg{\cdot}m^{-3}$ to 400 $mg{\cdot}m^{-3}$, resolution of ±0.1% for readings of 0.001 $mg{\cdot}m^{-3}$).

The sampling frequency for all monitoring devices was set at 60 s and each experiment was conducted in triplicate.

All instruments were calibrated by certified entities, which validated and demonstrated that every equipment was suitable for its intended purpose. For the Formaldemeter device, an in situ calibration was done before each monitoring period using a formaldehyde calibration standard supplied by the manufacturer, which takes into account the indoor temperature.

For the study of the burning emissions, the monitoring devices were placed at a distance of around 1.20 m from the source and at 80 cm from the floor. For the study of the cooking emissions, the monitoring devices were placed at a distance of one meter from the source. For the study of the toaster emissions, the monitoring devices were placed at a distance of 50 cm from the studied source. The selected location for positioning the equipment represented the point where the occupant spends more time and, consequently, where he/she is more exposed to pollutants. During all the experiments, only one person was present in the room where the activity under investigation was carried out. Monitoring devices were positioned at a minimum distance from the walls of 1 m and at a height corresponding to that of our nose, approximately, to avoid perturbations of the typical air flows, to minimize interfering emissions from other materials, and to represent the air of the breathing zone.

Before the start of each experiment, the studied room was well ventilated by opening windows and doors for 30 min and closing afterwards.

2.5. Statistical Analysis

A variance analysis of the results was performed using non-parametric statistics at a significance level of 0.050. All analyses were conducted using Excel software with the add-on XLSTAT 2014.1.09 [31].

3. Results and Discussion

Table 3 presents the IAQ guidelines established by the Portuguese legislation, which were considered to be limit values for the several pollutants addressed in this study.

Despite that the limit values refer to a daily average of eight hours, these thresholds will be used throughout the discussion of results to compare with the mean concentrations obtained for the different conditions under study, which lasted for a maximum of one hour and a half. It should be borne in mind that most domestic activities such as cooking or toasting are short-lived and do not allow the calculation of 8-h averages established in legislation. We must keep in mind that most domestic activities have a duration shorter than 8 h. Therefore, a small exceedance of the eight-hour limit value for a short-term activity may not necessarily be dangerous.

Table 3. Limit values of indoor air pollutants established by the Portuguese Ordinance no. 353-A/2013 [32] regarding a daily mean of eight hours.

Parameter	Limit Value
CO_2	2250 mg·m^{-3}
CO	10 mg·m^{-3}
VOCs	0.6 mg·m^{-3}
CH_2O	0.1 mg·m^{-3}
PM_{10}	50 µg·m^{-3}
$PM_{2.5}$	25 µg·m^{-3}

3.1. Fireplaces and Biofuel Type

Figure 5 depicts the results obtained for the four studied conditions. Table A1 of the Appendix A section presents the levels of all parameters for the period before combustion.

Overall, only VOCs and particulate matter were found to be above the limit values established by the Portuguese legislation [32] for some specific activities. Regarding comfort parameters, compared to background conditions (i.e., mean room temperatures ranging from 22.4 °C to 28.0 °C before combustion), all combustion scenarios provided increases in indoor temperatures from 18% to 24%. The combustion of briquettes in the fireplace insert promoted the highest increase of 24% on temperature, which corresponds to a mean rise of 6.5 °C. The use of pinewood in the same appliance led to the lowest mean increase (only 18%) among all the combustion scenarios, which corresponds to a mean rise of 5.0 °C.

The combustion of both biofuels in the open fireplace contributed to a similar room temperature increases of 5.6 °C and 5.5 °C for pinewood and briquettes, respectively. With regard to RH, a mean decrease ranging from 26% to 33% was registered depending on the combustion conditions. However, the recorded values were within the comfort range of 30% to 60% established by the international guideline ISO 7730 [33]. Due to the hot weather outside, the temperature values were above the established range for the occupant's comfort (23 °C to 26 °C for the summer period). Nevertheless, the magnitude of the increase of temperature or decrease of relative humidity was within the values reported by Salthammer et al. [19] who evaluated the impact of operating wood-burning fireplace ovens on IAQ.

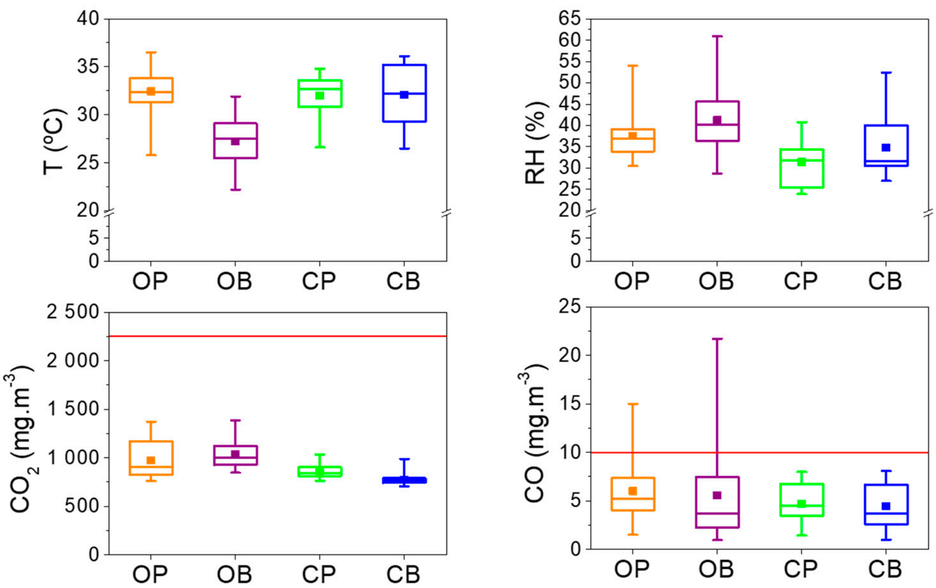

Figure 5. *Cont.*

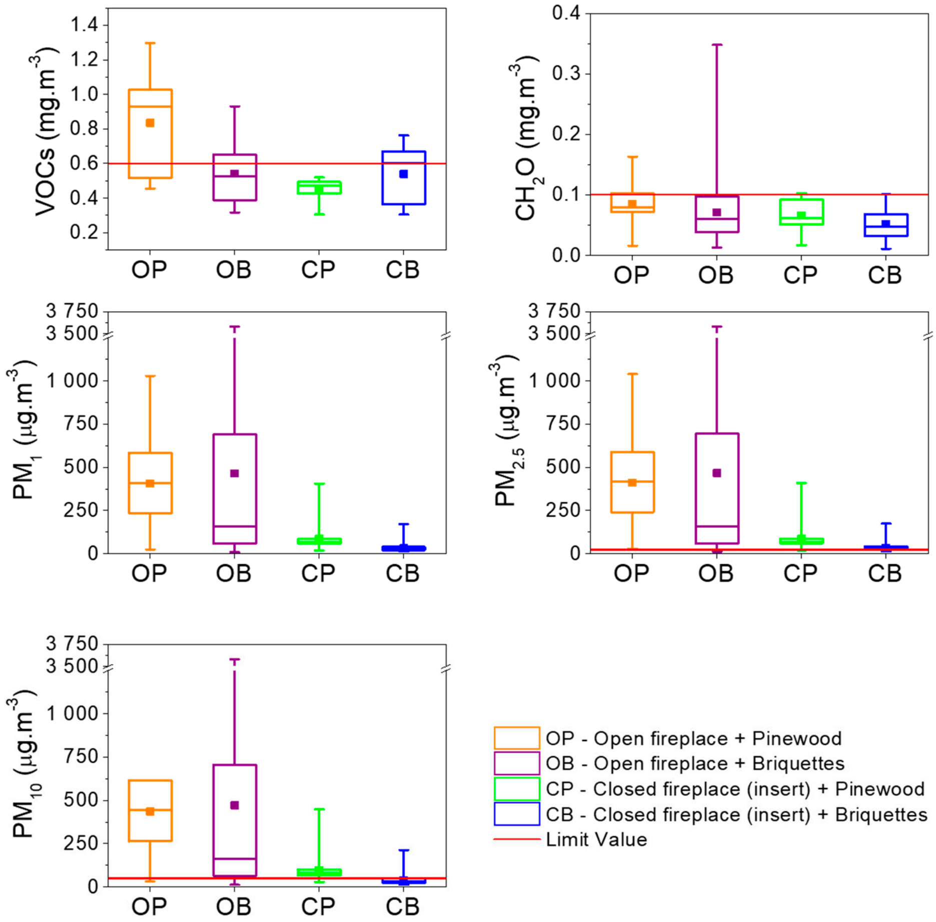

Figure 5. Indoor air pollutants and comfort parameters during the four combustion scenarios (OP—Open fireplace + Pine, OB—Open fireplace + Briquettes, CP—Closed fireplace (insert) + Pine, CB—Closed fireplace (insert) + Briquettes). The box plot represents mean (square) and the quartiles (5%, 25%, median, 75% and 95%).

3.1.1. Carbon Dioxide

For all combustion conditions, in general, CO_2 presented concentrations below the legislated limit value (2250 mg·m^{-3}), which is shown in Figure 6. The CO_2 levels in the living rooms remained relatively constant over the combustion cycles. However, it must be borne in mind that traditional combustion appliances such as open fireplaces or fireplace inserts are very inefficient. Most of the fuel carbon in wood is converted to CO_2 during the combustion process, but, because of the inefficient combustion, low combustion temperatures, and large amounts of excess air, a much higher ratio of carbon monoxide to CO_2 is produced in a fireplace burning wood than in modern wood-fired boilers. Incomplete combustion also leads to considerable emissions of particulate matter, VOCs, and other pollutants. Therefore, the relatively low CO_2 levels may be, at least in part, associated with inefficient combustion.

The highest CO_2 concentrations were found for the combustion of pinewood and briquettes in the open fireplace with mean levels of 974 $\pm$ 170 mg·m^{-3} and 1040 $\pm$ 140 mg·m^{-3}, respectively. Besides lower mean CO_2 levels, the combustion process in the fireplace insert contributed to smaller standard deviations (see Figure 6): 861 $\pm$ 57 mg·m^{-3} (pinewood) and 772 $\pm$ 38 mg·m^{-3} (briquettes). The uncontrolled and irregular intake of combustion air is the main cause that leads to higher standard deviations for the measurements carried out in the room equipped with an open fireplace.

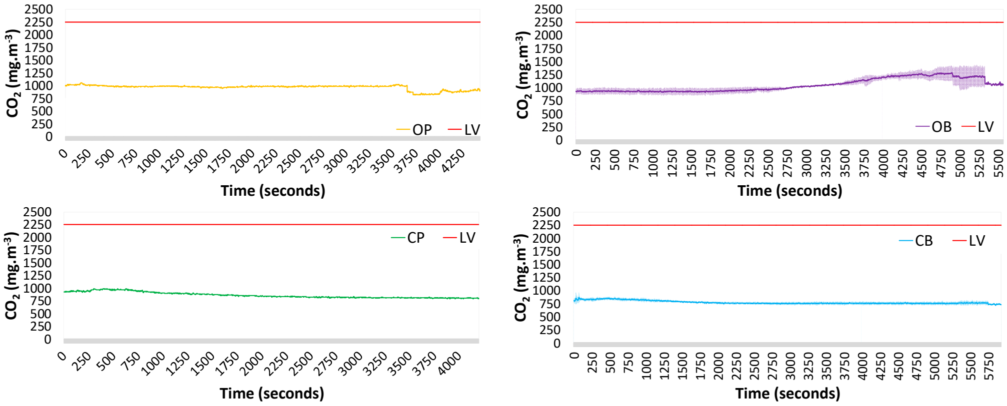

Figure 6. Temporal variability of CO_2 (line—mean; light colored area—standard deviation) during the combustion period for the four studied conditions: (**top**, **left**) OP—open fireplace and pinewood, (**top**, **right**) OB—open fireplace and briquettes, (**bottom**, **left**) CP—closed fireplace (insert) and pinewood, and (**bottom**, **right**) CB—closed fireplace (insert) and briquettes. Red line represents the CO_2 limit value of 2250 mg·m^{-3} defined by the Portuguese legislation [32].

3.1.2. Carbon Monoxide

The mean carbon monoxide levels were always below the limit value of 10 mg·m^{-3} set by the Portuguese legislation [32] (see Figure 7). However, the use of an open fireplace led to mean indoor CO values of 6.02 ± 3.33 mg·m^{-3} for pinewood and 5.55 ± 4.72 mg·m^{-3} for briquettes. In 32% (24 min) and 24% (22 min) of the combustion period for pinewood and briquettes, respectively, the limit value exceeded in at least one of the experiments. The CO mean values of this study are in agreement with the results reported by Salthammer et al. [19] who observed a half-hour mean indoor concentration of 5.11 mg·m^{-3} when operating an open fireplace. However, the results documented by the same researchers for closed fireplaces were always below 3.24 mg·m^{-3} while higher CO mean levels (4.71 ± 1.89 mg·m^{-3} and 4.43 ± 2.17 mg·m^{-3} for briquettes and pinewood, respectively) were registered in the present study. In Figure 7, it is possible to observe a sudden drop of the mean CO level without standard deviation for all combustion conditions. This mean CO level without standard deviation refers to one replicate of the experiment whose combustion period took longer than the other two and, therefore, no standard deviation was provided.

Carbon monoxide is a colorless and odorless gas. It is directly originated from incomplete combustion processes, which is often the case from fireplaces. Due to its affinity for hemoglobin, which is 200 times higher than that of oxygen [34], CO poisoning occurs after its inhalation. This reduces the blood's ability to carry oxygen and, therefore, blocks the oxygen to the body's organs and cells [35]. CO is the main cause of accidental intoxication, which leads to unintentional poisoning deaths in Western countries with an incidence of 23.2 poisonings per one million inhabitants per year in the United States [36]. From 1980 to 2008, in Europe, a total of 140,490 deaths by CO poisoning in 28 countries were registered with a mean annual rate of 2.24 deaths per 100,000 inhabitants [35]. From 2000 to 2007, a total of 621 hospital admissions by CO poisoning were registered in Portugal [37] with an incidence of 5.86 hospitalizations per 100,000 inhabitants per year. Colder months between November and March present peaks of incidence due to the use of house heating appliances that rely on inefficient combustion processes.

Taking into account that CO levels of 123 mg·m^{-3} are enough to trigger clinical symptoms [36], the levels found in this study for a maximum of 1.3 h of fireplace's use are below the alert value. However, a continuous use of these appliances for a longer period without proper ventilation may promote dangerous levels. Emissions greatly depend on both the combustion technology and operating conditions. With regard to human health, emission requirements for the eco-labelling of small-scale combustion appliances for wood logs and pellets should be mandatory in all countries. Strategies to decrease biomass burning emissions also include educational programs that advise people what to burn, how to burn, what are the less polluting appliances, etc. [38].

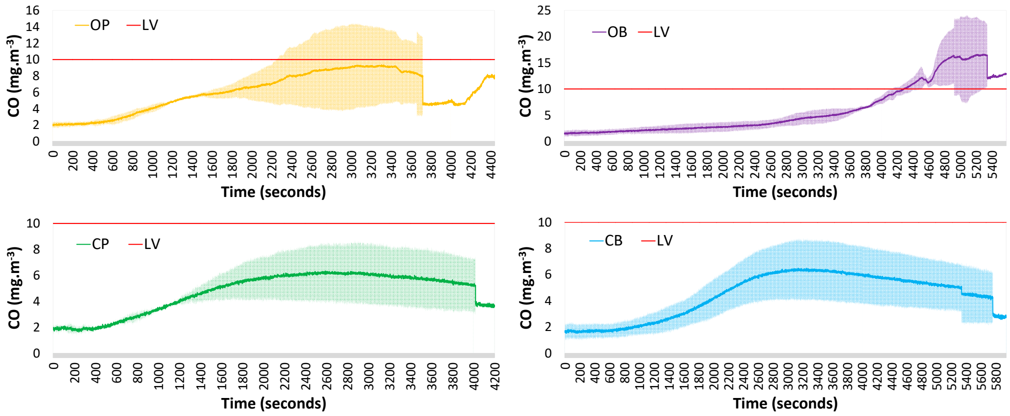

Figure 7. Temporal variability of CO (line—mean; light coloured area—standard deviation) during the combustion period for the four studied conditions: (**top**, **left**) OP—open fireplace and pinewood, (**top**, **right**) OB—open fireplace and briquettes, (**bottom**, **left**) CP—closed fireplace (insert) and pinewood, and (**bottom**, **right**) CB—closed fireplace (insert) and briquettes. The red line represents the CO limit value of 10 mg·m^{-3} defined by the Portuguese legislation [32].

3.1.3. Volatile Organic Compounds

Only the combustion of pinewood in the open fireplace led to a VOC mean level (0.836 $\pm$ 0.265 mg·m^{-3}) above the limit value of 0.6 mg·m^{-3} set by the Portuguese legislation [32]. The remaining combustion conditions contributed to mean values between 0.448 $\pm$ 0.054 mg·m^{-3} (combustion of pinewood in the closed fireplace) and 0.542 $\pm$ 0.173 mg·m^{-3} (combustion of briquettes in the open fireplace). However, some spikes above the limit value have been registered during the combustion process (see Figure 8).

The values of the present study are in agreement with those reported by Salthammer et al. [19]. These authors recorded VOC mean values from 0.18 mg·m^{-3} to 1.09 mg·m^{-3} regarding the burning emissions of different types of wood.

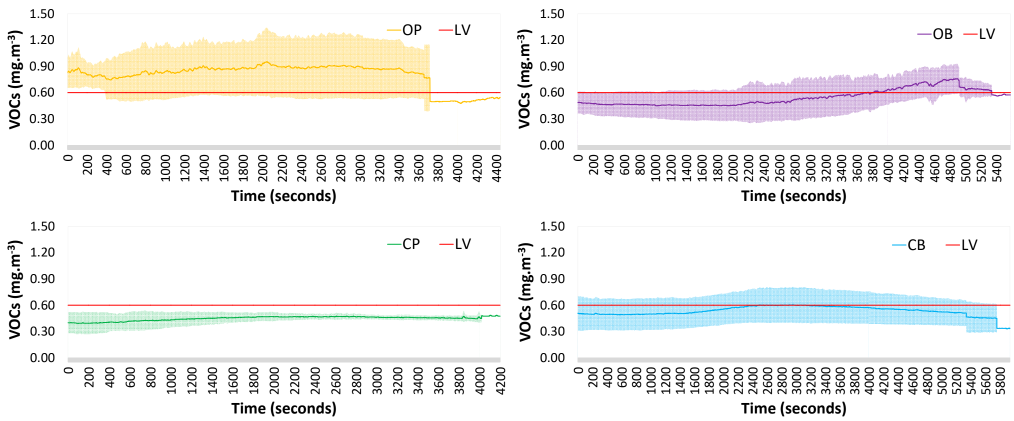

Figure 8. Temporal variability of VOCs (line—mean; light coloured area—standard deviation) during the combustion period for the four studied conditions: (**top**, **left**) OP—open fireplace and pinewood, (**top**, **right**) OB—open fireplace and briquettes, (**bottom**, **left**) CP—closed fireplace (insert) and pinewood, and (**bottom**, **right**) CB—closed fireplace (insert) and briquettes. The red line represents the VOC limit value of 0.6 mg·m^{-3} defined by the Portuguese legislation [32].

3.1.4. Formaldehyde

The mean CH_2O concentrations during all combustion cycles were below the national limit value of 0.1 mg·m^{-3} [32] (see Figure 5). The highest mean CH_2O concentration was found when the open fireplace was burning pinewood (0.085 ± 0.028 mg·m^{-3}) while the lowest mean levels (0.052 ± 0.024 mg·m^{-3}) were observed during the combustion of briquettes in the fireplace insert. However, some spikes throughout the combustion cycle in the open fireplace for both biofuels surpassed the limit value. Exceedances occurred for 17 min and 22 min when pinewood and briquettes were burned, respectively (see Figure 9).

It is noteworthy to refer that higher values of VOCs and CH_2O were found for the experiment involving an open fireplace and the combustion of pinewood (dwelling 2) for which the highest mean temperature, among all conditions, was reached (33.2 °C). The higher temperature in the room during the combustion period may have contributed to enhanced concentrations of VOCs and CH_2O because of higher desorption rates of these pollutants from furniture [39].

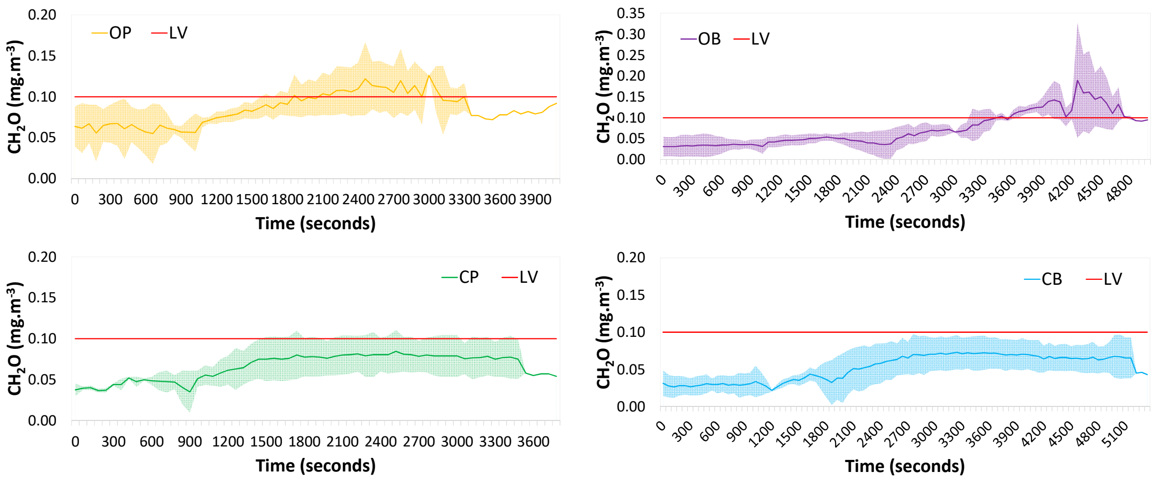

Figure 9. Temporal variability of CH_2O (line—mean; light colored area—standard deviation) during the combustion period for the four studied conditions: (**top**, **left**) OP—open fireplace and pinewood, (**top**, **right**) OB—open fireplace and briquettes, (**bottom**, **left**) CP—closed (insert) fireplace and pinewood, and (**bottom**, **right**) CB—closed fireplace (insert) and briquettes. The red line represents the CH_2O limit value of 0.1 mg·m^{-3} defined by the Portuguese legislation [32].

3.1.5. Particulate Matter

Figure 5 and Table 4 provide the results of particulate matter for the four studied conditions. The mean indoor levels of particulate matter throughout the combustion cycles were very high. Values below the protection limit of 50 µg·m^{-3} established by the Portuguese legislation [32] for PM_{10} were only achieved for the combustion of briquettes in the fireplace insert (39.3 ± 21.9 µg·m^{-3}). The use of pinewood in the same appliance promoted levels about two times higher than the limit value (96.9 ± 52.3 µg·m^{-3}). During the combustion of both biofuels in the open fireplace, PM_{10} levels were around nine times higher than the threshold.

The levels of $PM_{2.5}$ were always above the limit value of 25 µg·m^{-3} [32]. $PM_{2.5}$ accounted for 86% or more of PM_{10} with the combustion of briquettes in the open fireplace promoting the highest fine mass fraction (95% of PM_{10}). The combustion of briquettes in the fireplace insert also led to the lowest mean $PM_{2.5}$ concentrations (32.9 ± 17.2 µg·m^{-3}), which is slightly above the limit value. The use of pinewood in the fireplace insert promoted a higher mean value of $PM_{2.5}$ concentrations (86.0 ± 46.3 µg·m^{-3}), which were around three times higher than the limit value. However, for the open fireplace, mean $PM_{2.5}$ levels were 16 and 19 times higher than the protection limit of 25 µg·m^{-3} for pinewood and briquettes, respectively.

Table 4. $PM_{2.5}/PM_{10}$ ratios for the studied combustion settings.

	Combustion Conditions			
	Open Fireplace		Fireplace Insert	
	Pinewood	Briquettes	Pinewood	Briquettes
Ratio $PM_{2.5}/PM_{10}$	0.85 ± 0.14	0.95 ± 0.07	0.89 ± 0.07	0.86 ± 0.10

The time evolution of $PM_{2.5}$ concentrations resulting from the operation of both combustion appliances shows some differences, which is depicted in Figure 10. A decreasing trend in $PM_{2.5}$ levels is observed for the fireplace insert. This behavior reflects the combustion cycle with a first moment of high concentrations associated with the startup phase (ignition and devolatilisation) in which the door is open. This stage, is followed by a decay over time resulting from the closing of the door during the flaming phase, which leads to less infiltration of smoke into the room, and also from forced venting through the chimney.

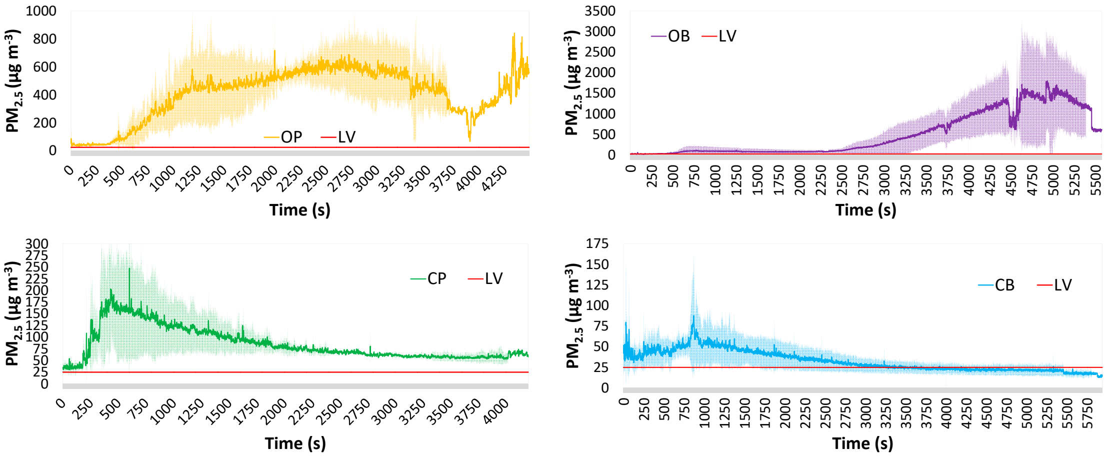

Figure 10. Temporal variability of $PM_{2.5}$ (line—mean; light coloured area—standard deviation) during the combustion period for the four studied conditions: (**top**, **left**) OP—open fireplace and pinewood, (**top**, **right**) OB—open fireplace and briquettes, (**bottom**, **left**) CP—closed fireplace and pinewood, and (**bottom**, **right**) CB—closed fireplace and briquettes. The red line represents the PM_{25} limit value of 25 $\mu g \cdot m^{-3}$ defined by the Portuguese legislation [2].

When comparing the PM levels before combustion (Table A1, Appendix A section), the biofuels used in the fireplace insert promoted the mean $PM_{2.5}$ and PM_{10} concentrations one (briquettes) and two (pinewood) times higher than the levels found before the combustion period. When the open fireplace was under operation, the PM levels were between nine and 20 times higher for PM_{10} (regarding pinewood and briquettes, respectively) and between 13 and 26 times higher for $PM_{2.5}$ (regarding pinewood and briquettes, respectively) than the PM levels registered during the period without combustion.

The particulate matter levels observed during the combustion of briquettes in the fireplace insert are in line with the mean values obtained by Salthammer et al. [19] in seven German private homes with different fireplaces (closed and open). A range from 6 $\mu g \cdot m^{-3}$ to 55 $\mu g \cdot m^{-3}$ was documented by these authors. A study in 50 homes (Montana, USA) [40] with wood stoves reported $PM_{2.5}$ concentrations of 32.3 ± 32.6 $\mu g \cdot m^{-3}$ (ranging from to 6.0 $\mu g \cdot m^{-3}$ to 163 $\mu g \cdot m^{-3}$) for a sampling period of 48 h. Results in the same range were obtained in a study in 96 homes (USA) [41] where, for two sampling periods, the mean daily concentrations of 28.8 ± 28.5 $\mu g \cdot m^{-3}$ and 29.1 ± 30.1 $\mu g \cdot m^{-3}$ were observed. When comparing particulate matter levels in living rooms of houses with central heating or wood heating [42], PM_{10} levels were found to be six times higher in houses equipped with wood heating systems (203 ± 201 $\mu g \cdot m^{-3}$) than in dwellings with central heating. Likewise, the $PM_{2.5}$ levels were found to be around 17 times higher in houses with wood heating (191 ± 193 $\mu g \cdot m^{-3}$) than in those with central heating.

It is known that emissions from residential biomass burning greatly vary with the type of biofuel, combustion equipment, and operating conditions [38]. In the present study, the use of open fireplaces promoted high concentrations of particulate matter in indoor air locations despite the type of biofuel used. Combustion of briquettes in a fireplace insert promoted the lowest PM concentrations while pinewood burning contributed to values similar to those monitored during the operation of an open fireplace. High levels of PM_X may have a negative impact on the occupant's health due to the submicrometric sizes and the presence of hazardous substances such as polycyclic aromatic hydrocarbons (PAHs) and oxygenated and nitrogenated derivatives, which have mutagenic and cytotoxic effects [43]. It has been observed that the combustion of conifer wood (e.g., pine) generates very intense flames, which are promoted by its high resin content. This leads to oxygen starvation and, consequently, to higher emissions of both particles and polyaromatics [27].

3.2. Cooking Emissions

Figure 11 provides an overview of the indoor pollutants for the two cooking processes (frying and boiling) using two different cooking appliances regarding the energy source (gas or electricity) in two dwellings.

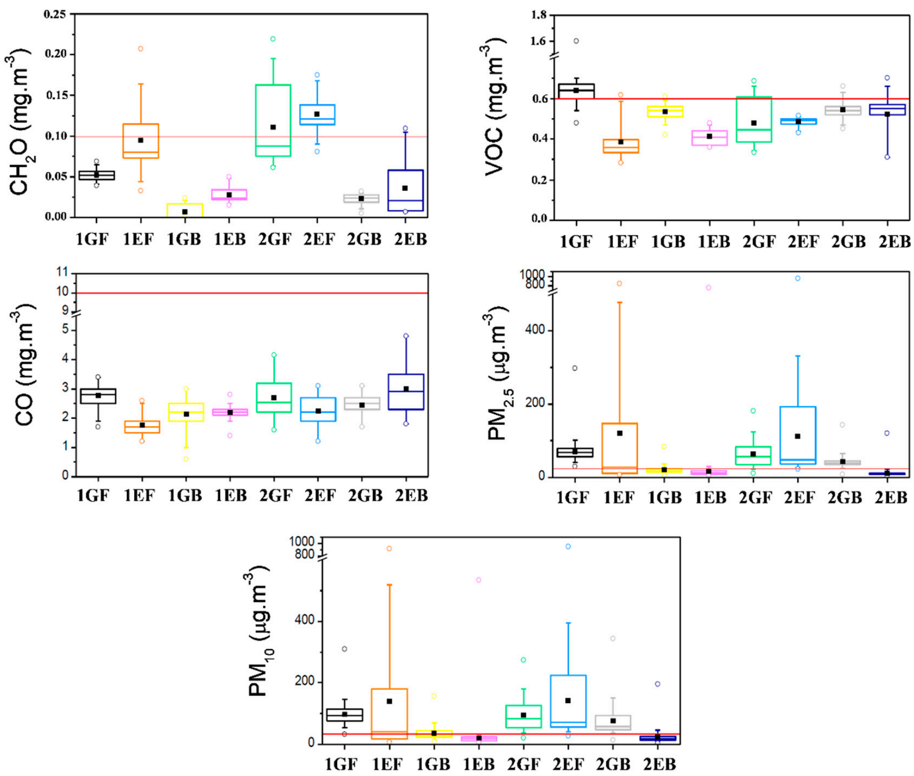

Figure 11. Selected indoor air pollutants for the studied cooking scenarios: 1GF—House 1 | Gas | Frying, 1EF—House 1 | Electric | Frying, 1GB—House 1 | Gas | Boiling, 1EB—House 1 | Electric | Boiling, 2GF—House 2 | Gas | Frying, 2EF—House 2 | Electric | Frying, 2GB—House 2 | Gas | Boiling, 2EB—House 2| Electric | Boiling, combustion scenarios. The red line represents the limit values of the Portuguese legislation [32]. The box plot represents mean (black square), the quartiles (5%, 25%, median, 75% and 95%), and lower circle the minimum value while the upper circle represents the maximum value.

Particulate matter levels (both $PM_{2.5}$ and PM_{10}) during the frying process were significantly higher than the ones found during the boiling process in both dwellings. Moreover, the mean concentrations during the frying process were above the protection limit established by the Portuguese legislation [32] for all experimental conditions (gas or electricity, both dwellings) for both fractions ($PM_{2.5}$ and PM_{10}). $PM_{2.5}$ and PM_{10} values above the limit value of 25 $\mu g \cdot m^{-3}$ and 50 $\mu g \cdot m^{-3}$, respectively, were registered when the cooking method was boiling using a device powered by bottled gas in dwelling 2. However, the levels were lower than the ones registered for the frying process. The difference in emissions between the two types of cooking methods (frying and boiling) agrees with the data presented by Abdullahi et al. [44] in their review paper, which shows that frying is among the cooking processes that promote higher exposure to particulate matter [45]. Cooking involving frying and grilling in kitchens was also shown by Nasir et al. [42] to lead to high fine PM concentrations, which reach mean maximum values above 1 $mg \cdot m^{-3}$. These values are higher than the ones observed in the present study.

Formaldehyde levels were found to be above the limit value of 0.1 $mg \cdot m^{-3}$ for the frying method at dwelling 2 (for both types of energy sources). At dwelling 1, the indoor levels when frying in an electric-powered device almost reached mean CH_2O values of 0.1 $mg \cdot m^{-3}$. VOC levels were only above the limit value of 0.6 $mg \cdot m^{-3}$ during the frying process with gas (dwelling 1) peaking at 1.6 $mg \cdot m^{-3}$ while, for the remaining cooking experiments, the values were below the threshold. Overall, VOC levels when cooking with the use of electricity were lower than the values when preparing meals with gas. Regardless of the cooking method or device, carbon monoxide was always below the limit value of 10 $mg \cdot m^{-3}$ established by the Portuguese legislation.

3.3. Emissions from Toasting

For the slightly toasted level, no value exceed the threshold, except for dwelling 2, in which the mean VOC levels was a little above 0.6 $mg \cdot m^{-3}$ (see Figure 12). Moreover, the results for particulate matter for slightly toasted agree with the ones described by He et al. [45] who reported a median $PM_{2.5}$ level of 35 $\mu g \cdot m^{-3}$ for the peak values, which are lower than the $PM_{2.5}$ levels of 62.6 $\pm$ 27.7 $\mu g \cdot m^{-3}$ and PM_{10} levels of 125.6 $\pm$ 87.1 $\mu g \cdot m^{-3}$ found for the toasting bread experiment in controlled conditions [18]. Both levels of medium and heavily toasted bread contributed to concentrations above the limit values for CH_2O, VOCs, $PM_{2.5}$, and PM_{10}. Concentrations increased with increasing toasting degrees with heavily toasting showing very high particulate matter emissions.

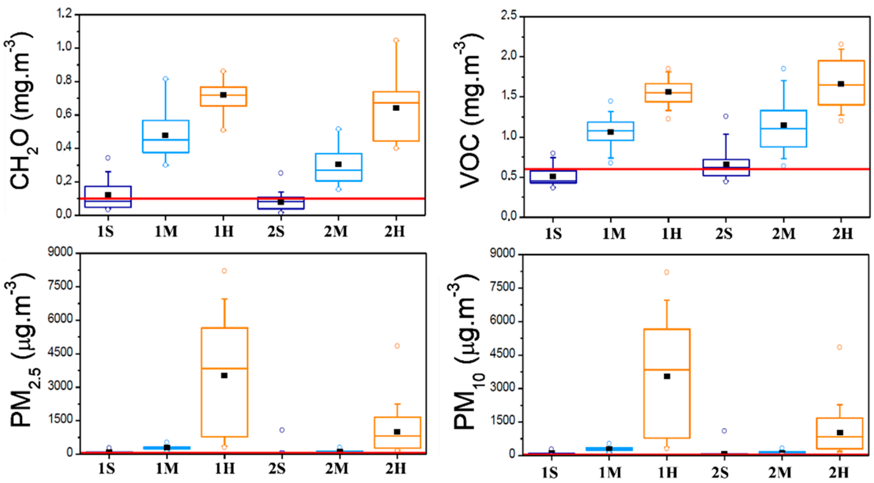

Figure 12. Indoor air pollutants for the studied toasting scenarios at two dwellings (1 and 2): S—slightly toasted, M—medium toasted; H—heavily toasted. The red line represents the limit values of the Portuguese legislation [32]. The box plot represents mean (black square), the quartiles (5%, 25%, median, 75% and 95%) and lower circle the minimum value while the upper circle represents the maximum value.

3.4. Discussion

The main limitation of the present study is the small number of measurements for each studied setting as well as the small number of dwellings studied. Although the selected dwellings represent the most common housing in Portugal, it is desirable in the future to increase the number of houses to confirm these preliminary results. Therefore, this study may be considered as the first approach toward understanding the effects of daily activities such as biomass home heating and cooking on the indoor air quality, which contributes towards assessing the exposure of occupants. Moreover, the evaluation of the dwellings' specific characteristics such as ventilation rates, airtightness, building's age and quality, and other factors have not been taken into consideration when selecting the studied buildings. To have a more robust understanding on how these factors may influence indoor air quality, it is desirable for future studies to gather a wider range of different dwellings along with a full characterization.

Despite that the location of the monitoring devices has been selected in order to simulate the occupants' exposure, only one measurement point per studied setting was defined. However, it is important to highlight that future studies should take into account the spatial variability within the same room in order to evaluate the likely concentration gradients of the pollutants.

In what regards standard deviations, it is normal to find a wide range of values over the whole period of activity given that the various phases show very variable emissions. It has been demonstrated, for example, that emissions of the ignition phase from a fireplace or stove may be one order of magnitude higher than those observed during the flaming phase. Concentrations spikes are also registered during the ash removal stage at the end of the combustion cycle [46]. The unequal behavior between gases (e.g., CO and formaldehyde) and particles may be determined by stove conditions that differentially influence the emission and dispersal of the various pollutants [47].

It is also important to stress the impact and contribution that carpeting and furniture may have on the indoor air quality due to different processes such as emission, adsorption, and desorption of pollutants from these materials [39,48]. This study focused on the worst ventilation conditions (e.g., windows and doors closed and hood switched off). It should be taken into account that the use of heating systems such as open fireplaces and inserts is commonly done in the wintertime. To prevent heat loss, owners often keep doors and windows closed. On the other hand, when cooking, the hood is often turned off especially because of the noise. In many homes, the exhaust system is only switched on when cooking is time consuming and emits intense odorous vapors such as the smells from fried foods. When a toaster is used, usually the hood is not turned on.

The promotion of ventilation and the use of exhaust systems are crucial for avoiding the accumulation of pollutants and improving the indoor air quality. This has been clearly demonstrated in a recent study in which the impact of different ventilation conditions on the indoor air quality during sleep was evaluated [13]. The condition with the window and door closed contributed most to the exceedances of limit values imposed for several pollutants (e.g., VOC, $PM_{2.5}$, and CO_2). Carbon dioxide levels were above the threshold below which the occupants showed significantly improved sleep quality in a study by Strøm-Tejsen et al. [49].

Despite this, users are not aware of the potential exposures of low ventilation practices and, in some activities, usually do not promote air exchange.

4. Conclusions

The present study allowed us to understand the impact of specific human activities in dwelling environments and their contribution to IAQ that occupants breathe. It was observed that activities such as the use of wood-fueled heating systems and cooking different types of meals (frying, boiling, and toasting) in different powered appliances may generate high levels of specific pollutants including VOCs, formaldehyde, and particulate matter, which can exceed the threshold values legally established. This is especially important given that frying fish sticks and chips as well toasting bread are widespread in many parts of the world including North America and Europe.

A fireplace insert compared to an open fireplace and briquettes compared to pinewood were found to contribute to lower indoor concentrations but still above limit values. The preparation of frying meals and heavy toasting in the kitchen promoted the highest levels of pollutants. Therefore, in order to minimize the occupant's exposure during these activities, it is crucial to promote natural ventilation and the use of efficient exhaust systems. Moreover, a careful selection of cooking equipment and fuels is also recommended to avoid risks. It is important to highlight that sometimes natural ventilation may lead to low air exchange especially in specific weather conditions. Therefore, the promotion of ventilation using natural or mechanical exhaust systems should always be considered as crucial for achieving a good indoor air quality in our dwellings especially when indoor emissions from human activities are taking place.

The main aim of the present study was to provide information with regard to users' exposure in the worst ventilation scenario. In the future, the same activities should be studied in different ventilation conditions in order to assess the variability in concentrations to which occupants are potentially exposed.

Author Contributions: S.M.A. and S.C. conceived and designed the experiments. S.C. performed the experiments. S.C. and N.C. analyzed the data. N.C., J.L., C.G., and C.A. wrote the paper. All authors have approved the final manuscript.

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

Appendix A

Table A1. PM_X, CO, VOCs, CH_2O, and CO_2 concentrations before the combustion period: mean ± standard deviation; median (minimum–maximum).

	Setting			
	Open Fireplace		**Fireplace Insert**	
Parameter	**Pinewood**	**Briquettes**	**Pinewood**	**Briquettes**
Temperature (°C)	27.6 ± 2.2; 28.8 (23.0–29.7)	22.4 ± 1.8; 21.8 (20.0–25.9)	28.0 ± 0.7; 27.5 (27.2–28.9)	26.5 ± 0.3; 26.5 (25.9–27.1)
Relative Humidity (%)	51.6 ± 6.3; 50.7 (44.2–66.2)	58.5 ± 7.4; 60.5 (45.8–71.3)	39.8 ± 0.7; 39.5 (38.7–41.1)	47.9 ± 4.1; 47.4 (42.0–54.6)
CO_2 (mg·m^{-3})	990 ± 186; 884 (794–1308)	949 ± 51; 973 (850–1155)	956 ± 45; 946 (913–1288)	812 ± 43; 820 (734–922)
CO (mg·m^{-3})	1.62 ± 0.56; 1.81 (0.00–2.42)	1.05 ± 0.54; 0.96 (0.00–2.35)	1.94 ± 0.13; 1.95 (1.43–2.29)	1.52 ± 0.41; 1.48 (0.71–2.40)
VOCs (mg·m^{-3})	0.838 ± 0.186; 0.898 (0.576–1.411)	0.486 ± 0.098; 0.471 (0.359–0.630)	0.399 ± 0.077; 0.345 (0.315–0.491)	0.538 ± 0.184; 0.519 (0.315–0.806)
CH_2O (mg·m^{-3})	0.056 ± 0.020; 0.053 (0.026–0.097)	0.029 ± 0.014; 0.021 (0.016–0.055)	0.040 ± 0.010; 0.044 (0.015–0.049)	0.031 ± 0.013; 0.025 (0.016–0.050)
PM_1 (µg·m^{-3})	34.7 ± 8.7; 32.0 (23.0–168.0)	19.5 ± 9.3; 14.0 (10.0–75.0)	34.3 ± 8.6; 37.0 (18.0–114.0)	34.5 ± 9.7; 37.0 (14.0–86.0)
$PM_{2.5}$ (µg·m^{-3})	38.5 ± 9.4; 36.0 (27.0–175.0)	21.1 ± 9.6; 16.0 (11.0–77.0)	35.8 ± 8.5; 38.0 (19.0–116.0)	37.4 ± 10.1; 40.0 (16.0–89.0)
PM_{10} (µg·m^{-3})	57.0 ± 23.6; 47.0 (30.0–267.0)	28.1 ±14.3; 22.0 (11.0–102.0)	56.4 ± 13.0; 55.0 (28.0–159.0)	64.8 ± 22.0; 57.0 (26.0–167.0)
Ratio $PM_{2.5}/PM_{10}$	0.72 ± 0.15	0.78 ± 0.11	0.65 ± 0.14	0.61 ± 0.17

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