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Determination of Pollutant Emissions from Wood-Fired Pizza Ovens

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The present study deals with the determination of pollutant emissions from wood-fired pizza ovens throughout the implementation of an innovative sampling system, specifically designed for the source in question. The experiments were conducted on a traditional wood-fired oven using two different forms of beechwood as fuel: wood logs and briquettes. Three different combustion cycles were used to carry out the tests, two for the wood logs and one for the briquettes. Different sampling sites along the system allowed for the collection and analysis of several gaseous pollutants, such as nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO_2) and OGC (organic gaseous carbon); along with total particulate matter (TPM). In order to study the synergistic effect of biomass burning and pizza cooking, combustion cycles were performed both with and without the presence of the pizzas. The results show that the concentrations of gaseous pollutants were affected mainly by the combustion conditions in the oven, whereas the values of TPM depended more on the amount of fuelwood used. Pizza cooking had a great impact on the emissions of OGC and, to a lesser extent, on CO and TPM.

1. Introduction

Non-industrial combustion is one of the eleven categories of emitters identified by the European Environmental Agency (EEA) listed in the Selected Nomenclature for Air Pollution (SNAP) (Dietmar, 1998). This macrosector includes combustion processes aimed at the production of heat for non-industrial activities such as commercial and institutional plants, residential heating and domestic combustion processes such as fireplaces and stoves, and agricultural installations (ARPA Sicilia, 2018). Non-industrial combustion contributes to the emissions of all major air pollutants (SO_2, NO_x, CH_4, CO, CO_2, N_2O, NH_3 and volatile organic compounds), with a significant impact on particulate matter. For instance, according to the most recent emission inventories developed by the INEMAR database, 42.5% of all the PM10 emitted in the Lombardy region of Italy can be traced back to non-industrial combustion, and this percentage reaches 49.1% if PM2.5 is considered (ARPA Lombardia, 2018).
Correlations between emissions from the civil sector and the different fuels used highlight the fact that wood combustion is the most relevant source of all major pollutants, with the exception of nitrogen oxides, in which case fossil fuels contribute the most (ARPA Sicilia, 2018). From the analysis of specific tracers such as levoglucosan and benzo(a)pyrene, several studies identified the rural mountainous areas as the most influenced by wood combustion; however, important contributions from biomass burning have been found also in urban areas (Lanzani, 2014). Moreover, the problem of air pollution deriving from wood burning is not confined to specific regions, but is widespread around the world, impacting mainly on the levels of PM2.5 (Favez et.al., 2009) and PM10 (Borrego et.al., 2010).
Wood burning in the non-industrial sector is employed mainly in small-scale domestic heating and cooking appliances including closed and open fireplaces, traditional and advanced stoves, boilers and kitchen stoves (Ozgen et.al., 2014), along with wood-burning ovens used in pizzerias and bakeries (Ielpo et.al., 2020). Whereas the contribution in rural mountainous areas is dominated by small-scale appliances used for heating purposes (Lanzani, 2014), in urban areas there is a non-negligible contribution deriving from wood-burning ovens (ARPA Sicilia, 2018). To the knowledge of the authors, there have been very few studies concerned with calculating the extent of this contribution. Within the Metropizza project (Hugony, 2018), a first attempt was made starting from emission inventory data using a modelling approach; the results showed that wood-burning pizza ovens in

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the province of Milan contributed in particular to the emissions of particulate matter and polycyclic aromatic hydrocarbons (PAHs).
Despite these preliminary studies, there is still an impressive lack of experimental data on the emissions of this source. To date, only one study has been published dealing with the characterization of particles emitted by ovens burning wood and briquettes. The results from this work highlight that wood-burning pizza ovens are an important source of fine particulate matter (PM2.5) characterized by a high content of black carbon (20-30%) (Mota Lima et.al., 2020). Moreover, there are no specific technical standards defining the constructional aspects of sampling systems or specifying tests methods for the analysis of pollutants. The only standard document that can deal with the subject is a 1995 UNI standard, currently withdrawn (UNI 10474:95), which deals with solid fuel ovens. In the broader field of biomass combustion, the European Commission has introduced source-specific emission standards in the Ecodesign Directive (EDD) for other appliances such as woodstoves and fireplaces, however excluding wood-fired ovens (Wolters, 2018).
The present work is a preliminary study on the emission profile of wood-fired ovens used in pizzerias. The aim is to determine the concentrations of the main air pollutants emitted by this source and to evaluate the extent to which pizza cooking impacts the overall emissions. In order to address these issues, an innovative sampling system was designed enabling the collection and analysis of the desired pollutants, and two different types of fuels were tested. Moreover, combustion cycles were created with the aim of reproducing the real-life operating conditions of the oven and at the same time ensuring repeatability and reproducibility of the measurements.

2. Materials and Methods

2.1 Appliance and fuels

The experimental tests were carried out using a traditional fixed top wood-fired pizza oven, which included a cooking surface made of refractory slabs with high-percentage alumina. A large amount of refractory material was also present under these plates, guaranteeing a supply of heat without temperature changes. Also, the cooking chambers were made of vibrated refractory concrete.
The oven was fed with beech wood logs normally sold to pizzerias, of length 50 cm, split diameter 6-8 cm, and with a moisture content below 20%. Instead, the wood briquettes were made of compressed beech wood without any use of adhesives or additives, certified for food use with a moisture content between 5.4-6.6%. The length of the briquettes was 30 cm and the diameter was 8.5 cm (octagonal shape).

2.2 Sampling system

The sampling system employed in the execution of experimental tests on the oven was constructed by making reference to the UNI EN 16510-1:2019 technical standard, which describes the test methods for sampling and measurement of the pollutant emissions of solid biomass residential heating appliances (Figure 1).
As reported in Annex F of the aforementioned standard, this system enables the simultaneous measurement of pollutants and other substances on the hot flue gases and on the cold flue gases after dilution in the tunnel. The dilution is controlled by a variable power exhaust fan located at the top of the dilution tunnel, and through a damper on a secondary section of the dilution tunnel.
The gaseous substances (CO, NO_x, CO_2, O_2) were measured both on the concentrated and diluted fumes, except for OGC which was measured only before dilution. Instead, TPM was sampled in continuous mode on the diluted gases using an opacimeter, and in discontinuous mode using appropriate filters on both the hot and cold flue gases. Also, the draught in the flue gas duct was continuously measured on the hot gases, and adjusted by varying the distance between the top of the measuring section and the suction hood of the dilution tunnel, as well as by varying the settings of the exhaust fan by means of an inverter.

2.3 Dilution factor

In order to report the concentrations of TPM measured in the diluted flue gases under the same conditions, it is necessary to determine the degree of dilution achieved in the tunnel. This was done by using the measured concentration values of CO_2 both upstream [$CO2_{upstream}$] and downstream [$CO2_{downstream}$] of dilution and subsequently applying the following formula to calculate the dilution factor, d, which takes into account also the concentration of carbon dioxide in ambient air, which was 300 ppm.

$$d = ([CO_{2_{upstream}}] - 0.03)/([CO_{2_{down\ stream}}] - 0.03) \quad (1)$$

Due to the fact that the dilution factor depends on the flue gas flow rate instantaneously generated by the combustion, as well as on the initial tunnel settings, this parameter may vary from test to test and during each test. It was therefore calculated punctually on the bases of the concentrations averaged over every measurement (every 10 seconds).

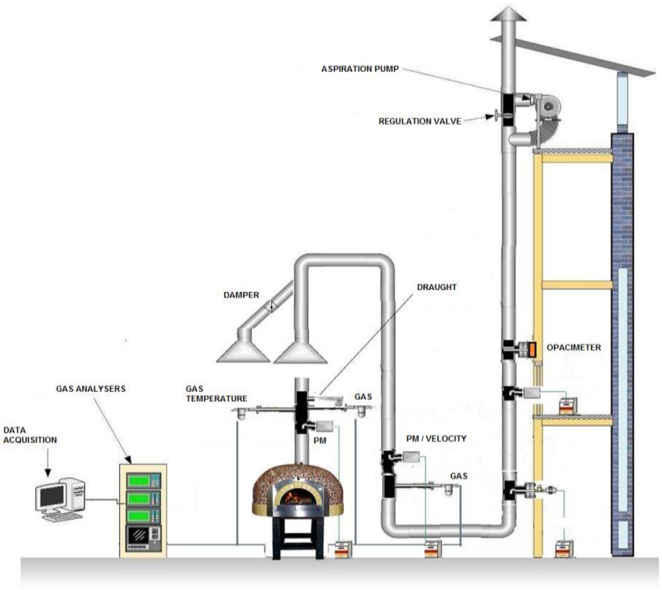

Figure 1: Scheme of the sampling system (not to scale)

2.4 Combustion cycles

In order to study the emissions of residential heating appliances burning solid biomass technical standards have laid out the combustion cycles that need to be followed (EN 13240:2001, EN 13229:2001, EN 14785:2006). Due to the lack of regulations regarding wood-fired ovens, new combustion cycles were developed specifically for the purposes of this study. Tests on wood-fired ovens must be conducted in such a way as to simulate, as far as possible, the real-life operating conditions of an oven in a normal public establishment. This is highly dependent on specific and local factors, such as the type of establishment and clientele, the season and local habits. However, it is necessary to define a standard reference cycle to be used in conducting the tests.
First of all, the oven used in the tests was managed following five operational phases:

1. Cold ignition: the cold furnace is ignited after several days of dormancy with a load of wood used as kindling, sold on the market specifically for this purpose;
2. Pre-heating: a series of fuel loads are selected for the tests and are fed to the furnace in sequence, taking advantage of the bed of embers formed during the ignition phase, in order to bring the furnace to operating temperature (approximately 350 °C);
3. Hot re-ignition: the furnace, which has been inactive for several hours (12-14), is re-ignited, as in point 1, when its internal temperature is still relatively high (80-180 °C), as is normally the case following the daily opening of a business, and is then fed with successive fuel loads until it reaches the operating temperature. At that point, the fuelwood and the embers are moved to one side of the oven, leaving space to cook the pizzas on the other side in the following phase;
4. Cooking phase: the hot oven is periodically fed with fuel loads in order to maintain a constant operating temperature, and at the same time, the pizzas are introduced. The normal operations of cleaning and moving the ashes and embers that are necessary for effective management of the oven under operating conditions are also carried out, but are excluded from the combustion cycles.
5. Shutdown: during this phase no fuel is supplied and no firing operations are carried out.

The pollutant emissions of the oven were measured during the cooking phase. The combustion cycles defined consisted of a single load of fuelwood, sampled for a fixed amount of time starting from the moment of entry of the fuel in the oven. Depending on the type of fuel and on the amount of beechwood used for the tests, different modifications were made to the standard reference cycle (Table 1).

Table 1: Combustion cycle details

Combustion cycle	Fuelwood	Mass of fuelwood supplied for each load / kg	Sampling time / minutes
A	Beechwood logs	1.7-1.9	20
B	Beechwood logs	0.8-1.0	15
C	Beechwood briquettes	1.7	15

In order to evaluate the impact of pizza cooking on the overall emissions, during each day of sampling 4 cycles were conducted with pizza cooking and 3-4 cycles were carried out without pizza cooking, for a total of 7-8 loads for each test on a particular fuel. The start of the first cycle of every day was set once the temperature of the oven reached 350°C; the others followed the first one.

2.5 Sampling methods and techniques

All the instrumentation used during the execution of the tests complies with the indications of the UNI EN 16510-1 technical standard and is managed in accordance with the specific reference technical standards for each parameter whose concentration is determined in the combustion fumes: UNI EN 14789:2017 for O_2, UNI EN 15058:2017 for CO, UNI EN 14792:2017 for NO_x, UNI EN 12619:2013 for OGCs, and UNI EN 13284-1:2017 for TPM.
All of the gaseous pollutants were measured using continuous analysers which were calibrated before the start of each test using calibration mixtures of known concentration. The fumes were extracted using a heated probe at 180°C and conditioned using a ceramic filter heated at 200°C, along with a Chilly compressor refrigeration unit and internal pump with dew-point output of 5°C.
Instead, smoke sampling for the determination of TPM was carried out on a section of the emission duct, as established by the reference standard, using a probe with a housing at the end in which the sampling filter was positioned; downstream of the probe are placed in succession: a smoke cooling device consisting of a condensate collector immersed in an ice bath, an absorption tower with silica gel and the suction pump.
The mass concentration of the emitted dust was determined gravimetrically after proper conditioning. The filters subjected to hot sampling were initially placed in an oven at 180°C for 1 hour, then in a desiccator for 4 hours, again in the oven at 160°C for one hour, and finally in the desiccator for another 4 hours before being weighed. Instead, the ones subjected to cold sampling were left to dry for 24 hours in a desiccator before being weighed.
Finally, the opacimeter used to sample TPM continuously on the diluted fumes was not calibrated: the absolute values do not reflect the actual dust concentration; the only purpose of this measurement was to highlight possible trends during the combustion cycles.

3. Results and Discussion

3.1 Pollutant emissions of the wood-fired oven

The main parameters kept under control for a correct execution of the test were: the draught, which significantly influences the combustion process and therefore the operation of the oven; the dilution factor, and the temperature of the diluted smoke at the sampling point, which affects the degree of condensation of semi-volatile organic compounds (SVOCs). Average values are reported in Table 2, along with other key sampling parameters.

Table 2: Sampling parameters (average values)

Combustion Cycle	Wood consumption / kg h^{-1}	Hot flue gas temperature / °C	Cold flue gas temperature / °C	Draught / Pa	Dilution factor
A	6.2	189	54.4	-18	5.4
B	5.1	153	44.1	-19	5.2
C	5.8	174	51.6	-16	4.8

Another important sampling parameter is the number of pizzas cooked during each day of tests: this was kept constant at a value equal to 16.
The emissions of the air pollutants for the various cycles were calculated as the average of the measured values for each cycle, with and without pizza cooking (Table 3). The impact of the pizzas will be discussed in the following section. NO_x and TPM concentrations fall within the wide range of values observed in literature data for domestic wood-burning appliances (Tomlin, 2021). Differently, when compared to the same devices, the

tested oven emitted lower concentrations of CO and OGC, closer to the performances of pellet fuelled appliances (Ozgen, 2014).

Table 3: Average concentrations of the measured substances

	HOT FLUE GASES						COLD FLUE GASES					
Cycle	NO_x / ppm	CO / ppm	CO_2 / %v	O_2 / %v	OGC / mg m^{-3}	TPM / mg Nm^{-3}	NO_x / ppm	CO / ppm	CO_2 / %v	O_2 / %v	Dust (opacimeter) / mg m^{-3}	TPM / mg Nm^{-3}
A	21.8	165	2.8	18.4	10.5	100 (±10)	4.3	34.2	0.54	20.5	30.0	91 (±8)
B	15.0	355	1.9	19.4	26.3	24 (±4)	2.8	71.8	0.39	20.8	6.0	30 (±3)
C	18.5	262	2.9	18.3	16.3	96 (±11)	4.2	59.3	0.66	20.3	36.1	105 (±9)

These results show that the concentration of CO and OGC emitted by the oven are greater when performing loadings using smaller amounts of beechwood separated by shorter time intervals (Cycle B). An opposite trend is observed in terms of NO_x and CO_2: these pollutants are emitted in greater amounts when performing loadings with greater amounts of beechwood logs separated by longer time intervals (Cycle A). This is consistent with the fact that emissions from biomass combustion are highly dependent on operational practices (Van Loo and Koppejan, 2008). Regarding Cycle C, the gaseous emissions of the oven were generally half-way between Cycle A and Cycle B, with the exception of CO_2, in which case the values were closer to Cycle A.
Based on experimental observations, loading more beechwood (Cycles A, C) enabled to reach higher average temperatures in the oven and keep a high flame throughout the entire sampling period. Instead, tests using Cycle B were associated with a reduction of the flame's intensity during the final sampling minutes, which caused the levels of CO and OGC to rise before the end of each cycle as a result of poor combustion conditions. Instead, no significant differences were observed in the trends of NO_x and CO_2 in all the aforementioned cycles. However, unlike other studies (Li et.al., 2012), higher temperatures were linked to greater emissions of NO_x.
With regards to TPM, Cycles A and C produced greater amounts of dust compared to Cycle B. This suggests that the amount of particulate matter generated is strictly related to the amount of fuelwood used. Overall, the comparison between TPM sampled on the hot and cold flue gases does not highlight a significant degree of condensation of semi-volatile organic compounds (SVOCs). On the one hand, this is in line with the findings of Ozgen et.al. (2014) which highlight a much lower difference between TPM concentrations on the hot and cold flue gases for open systems (open fireplaces, ovens) as opposed to closed ones (stoves, boilers). On the other hand, given that OGC was detected at concentrations above 10 mg m^{-3} in all three cycles, further measurements will be carried out to clarify this observation.

3.2 The impact of pizza cooking

Table 4 shows the average results obtained for experimental tests conducted with and without pizza cooking for the different cycles.

Table 4: Average concentrations with and without pizza cooking

		HOT FLUE GASES						COLD FLUE GASES					
Cycle	Pizza	NO_x / ppm	CO / ppm	CO_2 / %v	O_2 / %v	OGC / mg m^{-3}	TPM / mg Nm^{-3}	NO_x / ppm	CO / ppm	CO_2 / %v	O_2 / %v	Dust (opacimeter) / mg m^{-3}	TPM / mg Nm^{-3}
A	yes	22.4	182	2.9	18.3	13.7	109 (±16)	4.4	36.2	0.6	20.5	32.5	98 (±11)
	no	20.9	145	2.7	18.4	6.3	87 (±8)	4.2	31.4	0.5	20.5	26.6	75 (±6)
B	yes	15.5	354	2.0	19.2	35.3	31 (±1)	2.8	71.4	0.4	20.7	6.5	28 (±2)
	no	14.6	357	1.8	19.4	17.4	17 (±7)	2.9	72.3	0.4	20.8	5.5	26 (±5)
C	yes	18.6	284	2.9	18.3	24.3	93 (±16)	3.9	61.7	0.6	20.3	32.8	110 (±15)
	no	18.4	225	2.9	18.3	7.5	99 (±15)	4.1	50.2	0.6	20.3	29.5	100 (±11)

In terms of nitrogen oxides and carbon dioxide, no significant differences can be observed between the tests with and without pizza cooking. Differently, Cycles A and C suggest that this activity had an impact on the emissions of CO, whereas the results obtained for Cycle B do not show this trend, possibly due to large increases observed at the end of each trial which concealed the effect induced by the presence of the pizzas. Instead, the results from all three cycles agree on the fact that pizza cooking had a significant impact on the emissions of OGC. However, the gaseous compounds emitted from the pizzas did not have an impact on particulate matter. In fact, as already seen for the average results in the previous section, no significant amount of condensation can be observed in any one of the tests and the concentrations of TPM emitted with and without pizza cooking are not statistically different. One possible explanation could be that the expected contribution of the condensable fraction related to the higher OGC emissions associated with pizza cooking is concealed by the very high variability of the TPM measurements associated with wood burning. Once again, this problem will be further addressed in the following stages of this study.

4. Conclusions

From the results obtained in this study it is possible to conclude that wood-fired ovens represent an important source of major air pollutants. The concentrations of the examined species were comparable with the performances of other wood- or pellet-burning small-scale appliances. The experimental tests also showed that pizza cooking had a significant impact on the emission of OGC as a result of the volatilization of several compounds present in the food matrix.

Due to their extensive presence in many urban areas, the need to introduce regulation for this sector is of the upmost importance for the safeguard and improvement of air quality. At the European level, several actions have already been put in place in order to reduce the impacts of biomass combustion in the domestic sector, which could possibly be extended to include wood-fired ovens.

The next stage of the project will include the sampling and analysis of other air pollutants, such as polycyclic aromatic hydrocarbons (PAHs), with the aim to achieve a complete emission profile of this source. In addition, the contribution of PM2.5 and PM10 to TPM will be determined, along with a complete speciation of particulate matter into its various constituents: OC, EC, heavy metals, anions and cations. Furthermore, the emissions during ignition and shutdown of the oven will be established and compared with the ones determined during the cooking phase.

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