Characterization of Biomass Emissions and Potential Reduction in Small-Scale Pellet Boiler

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Abstract. In recent years it has been proved that residential biomass combustion has a direct influence on ambient air quality, especially in the case of cereals. The aim of this study is the characterization of the emissions in smallscale fixed-bed pellet boiler (heat output of 25 kW) of beech and corn, and of its potential reduction to an addition of calcium dihydroxide. In the biomass combustion test 7 fuel mixtures were investigated with regard to the particulate content (PM_{10}) , gaseous emissions and combustion chamber deposit.

The corn kernels tanned with calcium dihydroxide determined a decrease in particulate emissions $(54\pm13 \text{ mg } \text{MJ}^{-1})$ in comparison to corn, whereas in the combustion of corn pellet with 1% calcium dihydroxide high emissions were observed (193 \pm 21 mg MJ⁻¹). With regard to SO₂ emissions, the combustion of corn with the additives make a reduction in comparison to additive-free corn.

Keywords: Combustion, Emissions, Particulate matter, Beech, Corn, Pellet boiler.

1 Introduction

On average, biomass contributes some 9-10% to total energy supply in the world [1], but in European countries, biomass energy is 5-6% of total energy demand [2]. In Italy, a consumption of about 19 million tons of wood for domestic heating has been estimated (about 20% total energy consumed for residential heating) [3]. Wood is the most common and cheapest source of heat in many mountain areas during the winter, and it is burned in simple, tradition[al w](#page-14-0)ood stoves (with lower thermal efficiency) as well as in modern boilers (electronic control with the parameters). In recent years, however, a different possibility has become of interest, especially in rural areas (i.e. the use of agricultural residues such as straw, pruning of vines and fruit tree, residues of food processing, etc.).

Biomass combustion is regarded " $CO₂$ neutral", but at the same time is an important source of both gaseous and particulate pollutants (Table 1), such as particulate

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matter, carbon monoxide (CO), nitrogen oxides (NO_X) , sulphur dioxide $(SO₂)$ and hydrocarbons. The combustion emissions of biomass are associated with adverse health effects, such as pulmonary and cardiovascular symptoms [4, 5, 6]. Exposure to particulate matter has been estimated to cause an average loss of life expectancy of 9 months in EEA-32 countries (EU-25) [7]. As pointed out by Smith (1994), the products of incomplete combustion have three major adverse effects: energy loss, impact on human health and impact on the environment. He estimated that the use of biomass fuels contributed 1 to 5% of all CH₄ emissions, 6 to 14% of all CO emissions, 8 to 24% of all total non-methane organic compounds (TNMOC) emissions and thus 1 to 3% of all human induced global warming [8].

According to recent studies, small scale biomass combustion is the most important emission source of particulate matter in the cold season in Europe and in the USA. For example, particle emissions have been reported to be 50 times higher from a wood stove or a fireplace than from more controlled devices [9]. A survey conducted in five western Montana valley communities during 2006/2007 showed that wood smoke (likely from residential wood stoves) was the major source of PM_{25} in each one of the communities, contributing from 56% to 77% of the measured wintertime PM2.5 [10]. In 2010, an investigation in a residential site (Dettenhausen, Germany) concluded that during winter months the contribution from biomass combustion in the heating system to ambient particulate matter pollution was around 59% [11, 12]. Thus, it is evident that residential wood combustion has a direct influence on ambient air quality.

Fine particles originated in small-scale wood combustion mainly consist of ash, elemental carbon and organic material. Ash particles are formed when ash species volatilize in the hot combustion chamber and form particles when the flue gas cools down. In small-scale combustion, the most common ash particles are alkali metals, such as K_2SO_4 , KCl, K_2CO_3 and KOH [6].

Moreover, the combustion processes with biomass, especially with herbaceous biomasses which have a high content of alkaline elements, are prone to experience slagging, fouling and corrosion in the boiler plant. The biomass ash sintering on heat exchangers causes a decrease of heat transfer capacity and difficulty in cleaning the deposited ash. In relation to the heat exchangers, elements such as K, S and Cl are enriched in the ash deposits [13].

Recently, different studies have used additives into the biomass, in order to reduce the slagging phenomena in the plant and the particulate matter in the flow emission. In Denmark it is commonly recommended to add $1-2\%$ of limestone (CaCO₃) to avoid sintering [14]. Indeed, the combustion of cereal grains with the CaO in residential appliances (barely, rye and wheat) was shown to reduce slagging in a burner by formation of high-melting calcium/magnesium potassium phosphates. Thus, the formation of potassium fly-ash particles can be prevented by incorporation of potassium in calcium compounds they stay in the bottom ash [15]. Coda (2001) added some limestone to the straw combustion in a fluidised bed combustor, and observed that lime reacts with HCl, resulting in an enhanced share of Cl in the larger fly-ash particles, that was not bound as alkali chloride, followed by a corresponding reduction of gaseous HCl in the flue gas [16]. Subsequently, Wolf (2005) used Ca-based additives, including limestone, in high alkaline biomass combustion and found that emissions of gaseous SO_2 could be reduced with up to 25% [17]. It was suggested that limestone addition may produce a shift from alkali chlorides to alkali sulphates in the fine flyash particles. Recently, Bäfver (2009) concluded that the emission of particle mass can be lowered by supplying kaolin from residential combustion of oat grain (2% kaolin added to the fuel decreased the emission of particle with 31%, and 4% lowered the emission by 57%). On the other hand, however, the particle mass was not affected by addition of limestone, although limestone decreased HCl (Cl increased in the bottom ash) and $SO₂$ in the flue gas was unchanged because the high concentration of phosphorous in the fuel hindered $SO₂$ captured by limestone [15].

The objective of the present work was to investigate the effects of the biofuels mixture (corn with beech and corn with calcium dihydroxide $(Ca(OH₂))$) on the gaseous and solid emission in small appliances suitable for residential use. For the gaseous emissions were measured the total particulate matter and the concentration of the main pollution gases. The solid emissions were assessed in relation of the deposit in the combustion chamber.

Boiler/burner	Fuels	Power	CO ₂	\bf{CO}	CH ₄	NO _x	PM
		(kW)	$(\%)$	$(mg M J-1)$	$(mg M J-1)$	$(mg M J-1)$	$(mg MJ-1)$
Old-type boilers ⁽¹⁾ [9]	Wood	$6 - 24$	$4.7 - 8.4$	4.100-16.400	610-4.800	28-72	$87 - 2,200^{(5)}$
Modern boilers ⁽²⁾ [9]	Wood	$12 - 34$	$5.1 - 11.5$	507-3.781	$0.8 - 73$	$60 - 125$	$18-89^{(5)}$
Modern boilers ⁽²⁾ [9]	Wood pellet	$3 - 22$	$3.7 - 13$	$30 - 1.100$	$0-14$	62-180	$12 - 65^{(5)}$
Pellet boiler [5]	Wood pellet	25		80 ± 67	0.3 ± 0.2	49 ± 7	$19.7 \pm 1.6^{(6)}$
Stove (3) [4]	Beech	$6 - 6.5$		2.472-2.779	151-304.5	155-156	$111.4 - 131.3^{(7)}$
Stove (3) [4]	Oak	$6 - 6.5$		2.948-3.074	123.5-223.5	163-166	$107.3 - 121.8$ ⁽⁷⁾

Table 1. Range of emissions in the atmosphere from the combustion of various biomass, expressed as mass per energy [4, 5, 9]

(1) Water-cooled multi fuel boiler, up-draught combustion

(2) Ceramic wood boiler with fuel gas fan, down-draught combustion

(3) Logwood stove with manual operation

(4) Improved biomass stove for space heating in developing countries

(5) Mass of particles below aerodynamic diameter of 2.5 μm

(6) Mass of particles below aerodynamic diameter of 1 μm

(7) Mass of particles below aerodynamic diameter of 10 μm

Nomenclature

Feedstock

- C Corn grain
- CL Corn grain tanned with 1% calcium dihydroxide
- CB12 Pellet of corn (87,5%) and beech (12,5%)
- CB25 Pellet of corn (75%) and beech (25%)
- CB50 Pellet of corn (50%) and beech (50%)
- CFL Pellet of corn (99%) and calcium dihydroxide (1%)
- B Pellet of beech

Symbols

2 Materials and Methods

The experiments were carried out to assess the emissions of the main flue gases (O_2) , CO_2 , CO , NO , NO_2 , NO_X and SO_2) and particulate matter in a fixed-bed boiler with a maximum heat output of 25 kW.

The experiments were performed with seven combinations of biomass samples from agro-forestry products (Table 2) and additives, added to improve the combustion and reduce emissions of pollutants in the atmosphere. For each sample were made three tests. The feedstocks included:

- corn grain and beech pellet;
- pellet of corn and beech $(12.5\%, 25\% \text{ and } 50\%);$
- pellet of corn and 1% calcium dihydroxide $(Ca(OH)₂)$;
- corn grain tanned with calcium dihydroxide $(Ca(OH₂)).$

The pellet samples were obtained by mixing corn flour with ground beech and/or calcium dihydroxide additive. The mixture was pelletized in a professional installation (Costruzioni Nazzareno s.r.l., Breda di Piave (TV) - Italy). The tanned corn samples were obtained by mixing whole corn with the additive (calcium dihydroxide), and then manually sieving the mixture to remove the excess additive. The percentage of additive was determined base on the weight difference before and after treatment (0.87%). The test did not include pellet samples made with pure corn flour since, because of technical and economical aspects, it is hardly convenient to pelletize the flour instead of using the whole corn directly.

Table 2. Characteristics of biomass samples used in the tests, and operating parameters of the appliances

U, in %, is the biomass moisture content

LHV, in MJ kg^{-1} d.b, is the lower heating value on dry basis

EC, in $MJ kg^{-1}w.b.,$ is the energy content on wet basis

P, in W, is the nominal heat input

B, in kg h^{-1} , is the burning rate

 F , in $m³ s⁻¹$, is the flow of the gas emission

The lower heating value (LHV) of C and B was determined with a calorimetric bomb (C200, Ika, Staufen Germany); for the other sample feedstocks, the energy content was calculated based on the percentage of the different components. Afterwards, the energy content was calculated taking into account the latent heat of the water absorbed in the structure of the biomass. The following formula was used to calculate the energy content [19]:

$$
EC = \frac{LHV \cdot (100 - U) - 2.44 \cdot U}{100}
$$
 (1)

where: EC, in MJ kg^{-1} w.b., is the energy content; LHV, in \overline{MJ} kg⁻¹d.b., is the lower heating value; U, in %, is the moisture content of the biomass; 0.025, in MJ kg^{-1} , is the heat of water vaporization.

Table 2 shows the analysis of moisture and energy content of the feedstocks. The combustion equipment was similar to most commercial pellet appliances (Granola 25R, Arca Caldaie, San Giorgio di Mantova - Italy) with slight differences in the hydraulic and heat dissipation systems (Fig. 1).

The small-scale pellet boiler represented a modern continuous combustion technology with a nominal power output of 25 kW and can be operated with any kind of wood and herbaceous feedstock, either in form of pellets or grain. In this combustion equipment there is not a hot water cylinder (buffer tank) as a heat storage and the heat is directly dissipated by a fan heater.

Fig. 1. Combustion equipment: a) fuel tank, b) screw feeding, c) combustion chamber, d) turbulators for heat exchangers, e) steel chimney (12 mm), f) fan heater, g) boiler control unit

The feedstock is supplied to the combustion chamber from an external fuel storage by a transport screw. The chamber combustion had a square shape with a pig iron plate for gas circulation. Its size was 520 mm, 550 mm, 590 mm (width, length, height). The fumes pass to the heat exchanger of the boiler and subsequently in the steel chimney, where the sampling probes were placed.

Biofuel boilers typically present a very long period of start-stop, and it takes them a long time to work at full performance. For this reason, the biomass plant included a heat sink, whose function was a thermic dissipation, that allows to maintain a constant power of the combustion. The biomass combustion was corrected with the operation time of the screw feeding and the flow air with the electronics unit (Termotre 32.10, Tecnosolar, Villimpenta (MN) - Italy). In this way, it was possible to obtain a constant power (about 20 kW) and determine the fuel consumption for the all test (Table 2). The tests performed during biomass combustion have provided the following samplings (Fig. 2):

- the flue gas emission was measured with a multi-gas analyzer (Vario plus industrial, MRU air Neckarsulm-Obereisesheim; DIN EN 50379-1+2). Flue concentrations of CO_2 and CO were measured with an infrared bench, while O_2 , NO, NO₂ and SO2 were measured electrochemically with a sensor with three electrodes. The NO_x gas was calculated as the sum of NO and $NO₂$;
- the particulate matter was assessed with a gravimetric method, which required the isokinetic conditions to collect the particulate (UNI EN 13284-1 and UNI 10169). The gas emission was collected with a constant flow rate sampler (ZB1 battery, Zambelli, Milano - Italy). The speed of flue gas was measured with a Pitot tube connected to the multi-gas analyzer and the flow of the sampling probe was corrected (gas speed per 6 mm diameter probe). The particulate matter was collected on quartz fiber filters with a 47 mm in diameter (FAFA quartz filter, Millipore, Billerica Massachusetts – USA). Its efficiency is >99.998% (0.3 micron particles) and it is considered the most penetrating particle in an air flow stream (maximum temperature of 950 \degree C);
- the temperature in the combustion chamber was measured using a thermocouple (Tastotherm MP 2000, Infrapoint, Saalfeld – Germany);
- the deposit in the boiler was measured with a three steel plates in the combustion chamber, before the heat exchanger (40 mm x 25 mm x 8 mm).

The experimental analyses were carried out in the summer when the ambient humidity was lower than 65% and temperatures varied between 20 and 32°C. For all tested boilers, the same methodology was performed. Initially, the boiler was turned on and after 30 minutes and not before the chamber temperature was stable, the emissions were recorded for 60 minutes. The gas emission values were measured in parts per million (ppm, volume to volume) and then expressed as a mass of pollutant per content energy of the fuel $(mg MJ⁻¹)$. The following formula was used to calculate the pollution mass to volume concentration:

$$
C_{m/v} = C_{v/v} \frac{FM}{22.4} \quad (2)
$$

where:

 $C_{m/v}$, in mg m⁻³, is the mass to volume concentration; $C_{v/v}$, in ppm, is the concentration of volume to volume; FM , in g kmol⁻¹, is the molecular weight; 22.4, in m³ kmol⁻¹, is the specific volume of a gas at 20 $^{\circ}$ C temperature and 101.325 kPa (Avogadro's law).

The following formula was used to calculate mass pollution to biomass energy:

$$
C_{m/e} = \frac{C_{m/v} \cdot F}{P \cdot 10^6} \qquad (3)
$$

where:

 $C_{m/e}$, in mg MJ⁻¹, is the mass per unit of biomass energy;

 $C_{m/v}$, in mg m⁻³, is the mass to volume concentration;

F, in m^3 s⁻¹, is the flow rate of the gas emission (gas speed per 12 mm steel chimney diameter);

P, in W, is the nominal heat input.

Fig. 2. Schematic of the experiment set-up: a) steel plates in the combustion chamber, b) pitot tube; c) instrument probe for combustion gas sampling, d) probe for dust sampling, e) multi-gas analyzer; f) silica gel to remove moisture from the flue gas; g) dust sampler; h) instrument to measuring the temperature in the combustion chamber; i) computer for data collection

For the particulate matter, the quartz filters were collocated after the analysis in a heater at 105^oC, in order to remove residual moisture. The filters were subsequently weighed to determine the total particulate matter collected. The following formula was used to calculate the PM to biomass energy:

$$
PM = \frac{M_{\rm m}}{P \cdot 3,600 \cdot 10^6}
$$
 (4)

Where:

PM, in mg MJ^{-1} , is the particulate mass per unit biomass energy; M_m , in mg, is the mass of particulate matter collected by the filter; P, in W, is the nominal heat input; 3,600, in s, is the duration of the test.

The plates were weighed before and after the experimental test, in order to determine the amount of dust deposit. The weight was related to the mass of deposit per unit energy (mg m^2 MJ⁻¹) in the fuel (Fig. 2). The following formula was used to calculate the mass deposit to biomass energy:

$$
D_e = \frac{D_m}{SP \cdot 3,600 \cdot 10^6} \qquad (5)
$$

where:

De, in mg m^{-2} MJ⁻¹, is the mass deposit per unit sampling area in relation to biomass energy;

Dm, in mg, is the total mass deposit on the plates;

SP, in m^2 , is the surface of steel plates;

P, in W, is the nominal heat input;

3,600, in s, is the duration of the test.

3 Results and Discussion

The results concern O_2 , CO_2 , CO , NO_2 , NO_2 , NO_2 , SO_2 , PM and De . The total gaseous and solid emissions (amount of emitted substance per MJ of energy in the fuel) of these biomass fuels are also given in Table 3 and in Table 4.

Data showed higher emission of CO (unburnt pollution) in the flue gas from the beech wood pellet, in comparison to the corn. As for pellet of corn with beech the unburned emission were similar to beech combustion. In relation to the additive utilization the corn grain tanned with $(Ca(OH)_2)$ make a decrease of CO in comparison to corn combustion (-32%) while the pellet combination with corn and 1% (Ca(OH)₂) gave an increase of CO (+158%) (Table 4). The analysis showed a strong variability of CO emissions (Fig. 3). It has been observed that CFL combustion results were more variable than all the others analyzed samples. Perhaps, calcium adsorbs the heat in the combustion chamber and causes a strong variability in the combustion temperature $(795^{\circ}C \pm 10)$ [22-28].

	Symbol	\mathbf{O}_2	\mathbf{CO}_2	PM	Deposit	TE	ТC
Feedstock		$(\%)$	$(\%)$	$(mg M J^1)$	$(mg cm-2 m-3)$	$(^{\circ}C)$	$(^{\circ}C)$
Corn grain	C	15 ± 0.1	5.7 ± 0.1	74 ± 9	12 ± 8	140.0 ± 0.3	734 ± 6
Corn grain + 1% hydrated lime	CL.	14.6 ± 0.1	6.1 ± 0.1	54 ± 13	108 ± 21	151 ± 0.6	613 ± 8
Corn flour (87.5%) + Beech $(12.5%)$	CB_{12}	14.9 ± 0.0	6.1 ± 0	$72 + 1$	$13 + 6$	152.8 ± 0.3	$680 + 7$
Corn flour $(75%)$ $+$ Beech $(25%)$	CB ₂₅	14.6 ± 0.0	6.3 ± 0.1	65 ± 4	14 ± 5	155.7 ± 0.2	$726 + 7$
Corn flour (50%) $+$ Beech (50%)	CB_{50}	15.6 ± 0.1	5.1 ± 0.1	66 ± 9	$8 + 5$	153.6 ± 0.5	804 ± 8
Corn flour (99%) + 1% hydrated lime	CFL.	14.4 ± 0.0	6.3 ± 0	$193 + 21$	$62 + 2$	162.2 ± 0.3	795 ± 10
Beech	B	15 ± 0.1	5.6 ± 0.1	164 ± 11	15 ± 1	158.0 ± 0.2	$751 + 5$

Table 3. Emissions of O_2 , CO_2 , PM, $C_{d/v}$, TE and TC of fuel combustion (the standard error is reported to the average of the individual repetitions for each sample)

PM is the particulate matter

TE is the temperature of the flue gas emission

TC is the temperature in the combustion chamber

Fig. 3. Box plot of the variability in the CO emission for the all tests (in the simplest box plot the central rectangle spans the first quartile to the third quartile; the segment inside the rectangle shows the median and above and below the box show the locations of the minimum and maximum).

One of the main environmental impacts of solid biofuel combustion is caused by NOX emissions. The main mechanism of NOX formation in the fuel biomass, when the temperature of combustion is between 800-1,200°C, is the oxidation of the nitrogen contained in the fuel. The determining factors are the amount of fuel bound nitrogen in the ultimate analyses, the O2 concentration in the flame area and in a lower degree its temperature [22]. Furtermore the geometric of furnace and the type of combustion technology applied influencing variables for NOX formation [21]. According to Table 4, beech wood combustion have the lowest nitrogen oxides emissions (145±2 mg MJ-1), while higher concentrations are found in corn (447±4 mg MJ-1) and in corn mixed with beech (about 414 mg MJ-1). The emission data confirm the higher nitrogen content in the fuel, the higher NOX emissions from the biomass combustion.

Table 4. Emission of CO, NO, NO₂, NO_x, and SO₂ of fuel combustion (the standard error is reported to the average of standard error of the individual repetitions for each sample)

HC is the hydrocarbon combustion expressed as CH4

Fig. 4. Box plot of the variability in the NO_X emission for the all tests

The emissions of NOX for the combustion of corn grain tanned with calcium dihydroxide $(412\pm9 \text{ mg MJ-1})$ resulted the same of corn combustion whereas the pellet combination with corn and 1% calcium dihydroxide has resulted in an increase of NOX emissions (566±7 mg MJ-1). The variability of NOX emissions resulted to be very high for CL, CLF, CB25 and CB50 (Fig. 4).

The sulphur contained in the solid biofuel forms mainly gaseous SO2. Due to the sub-sequent cooling of the flue gas in the boiler section of the combustion plant, SOX forms sulphates and condenses on the heat exchanger surfaces or reacts directly with

Fig. 5. Box plot of the variability in the SO_2 emission for the all tests

Fig. 6. Emissions of particulate matter (PM) and deposit (Cd/e) in combustor chamber for the feedstock combustion (reported the standard error of the repetitions for each samples)

Table 5. List of the equation in the text

fly ash particles deposited on heat exchanger surfaces (sulphation). The average emission of sulphur dioxide in corn combustion were 67 ± 2 mg MJ-1 while beech wood emissions were lacking because these fuels has a low concentration (Table 4)

The emission for corn with beech was smaller in comparison to corn (-72% for CB12, -31% for CB25 and -87% for CB50). The efficiency of sulphur fixation in the ash depends on the concentration of alkali and earth-alkali metals (especially Ca) in the fuel (fuels like wood chips and bark can have high Ca contents and cause therefore a high S fixation) [20, 21]. This consideration was confirmed by the result of the lime utilization; the emission were lower by 31% for CL and 48% for CFL.

The mass concentration of particulate matter from combustion of corn and pellet corn with beech ranged from 65 to 74 mg $MJ⁻¹$. The highest concentration occurred during the combustion of beech wood with 164 ± 11 mg MJ⁻¹. The utilization of lime as an additive in the corn pellets resulted in increased emissions of particulate matter $(193\pm21 \text{ mg } \text{MJ}^{-1})$ while the use of lime tanned with corn resulted in a decrease of emissions 54 ± 13 mg MJ⁻¹). Probably this decrease in emissions was determined by the dust of calcium dihydroxide in the combustion chamber that captured particulates matter (Fig. 6). The deposit in combustion chamber was the same for the feedstocks without the additive although there was tendency to lower emission for the CB50 (- 33% in comparison to corn combustion) [29-31]. In relation to the additive utilizations, shows a significant increase of the deposits in the combustion chamber caused by the presence of calcium compounds formed during combustion (+800% for the CL and +417% for the CFL in comparison to corn combustion).

4 Conclusions

The results presented in this study clearly show that substantial differences in the combustion emissions were produced by the different feedstock from small-scale appliances. The unburned (CO and PM) emitted from combustion of corn was smaller than the emission from beech combustion, but the NO_X emission was much higher for corn utilization because there is higher nitrogen content in the feedstock. The calcium dihydroxide added to the fuel in the corn pellets reduces the $SO₂$ emissions. Conversely the additive increase the emissions of particulate matter and NO_X emissions from corn feedstock.

The calcium dihydroxide may be used to reduce the corrosive gasses, but the operation of the boiler and the method of the additive supplied play an important role for the result. In the future experiments, more focus should be put on the residence times of the fuel in the combustion chamber and the reaction with the bottom ash. In additional, a set of chemical analysis is recommended to be performed the particulate emissions and the bottom ash in order to provide more specific data to aid in the evolution of reduction emissions to small-scale biomass combustion.

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