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1 Gaseous and Particulate matter emissions from the combustion of biomass-based insulation  
2 materials at end-of-life in a small-scale biomass heating boiler

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### 9 **Abstract**

10 To alleviate the unbearable pressure on forests in Europe, the use of lower quality woody  
11 materials or wastes and agricultural fuels is emerging for the production of heat. The two major  
12 challenges of using these types of fuels are associated with the chemical composition variability  
13 and some fuel characteristics that could trigger potential issues throughout the burning process.  
14 This study aimed to investigate the environmental and technical performance of a small-scale  
15 multi-fuel biomass heating boiler (20 kW) fired with DIN*plus* certified wood pellets and two  
16 granulated biomass-based insulation materials, namely hemp fibers and wood fibers under  
17 standard laboratory conditions. The boiler efficiency, particulate matter, and gaseous emissions  
18 were evaluated and compared to the legally permissible values specified in EN 303-5:2021.  
19 Hemp fiber pellets fulfilled the requirements for residential pellets, while wood fiber pellets  
20 showed lower durability and higher nitrogen and sulfur contents than the European standard  
21 limits. To respect the emissions regulation, blending wood fibers with certified wood is  
22 therefore necessary. Pellets type was noticed to significantly affect the gaseous and dust  
23 emissions but marginally the boiler efficiency. This latter was near 80% for all fuels owing to  
24 high sensible and latent heat losses. Hemp fiber pellets showed high CO and dust emissions

25 attributed to their high bulk density and high ash content, respectively, while the wood fiber  
26 pellets showed high NO<sub>x</sub> and SO<sub>x</sub> emissions accredited to their elemental composition. Gaseous  
27 and dust emissions were reduced through the blending of hemp and wood fibers with certified  
28 wood pellets. The particulate mass concentrations of the tested pellets fluctuated between 38  
29 and 360 mg.Nm<sup>-3</sup>. Particulates were mainly composed of potassium, chlorine, sulfur, silicate,  
30 calcium, and zinc. From a number perspective, particles with diameters below 1 μm were  
31 dominant and their concentration varied between 10<sup>4</sup> and 10<sup>6</sup> particles.cm<sup>-3</sup>. Pelletized biomass-  
32 based insulation materials mixed with certified wood could be considered an attractive  
33 alternative fuel for residential biomass boilers.

34 **Keywords**

35 Biomass-based insulation materials, Boiler efficiency, Combustion, Gaseous emissions,  
36 Particulate matter, Pellets

## 37 1. Introduction

38 Rising energy consumption and fossil fuel prices, growing environmental awareness  
39 regarding their use and emissions, in addition to concerns about the security of energy supply  
40 have fostered the research for renewable energy sources around the world [1,2]. In 2017, the  
41 share of these renewable energies reached 13.9% of the total primary energy supply and 17.7%  
42 of the gross final energy consumption (GFEC) worldwide [3]. Globally, biomass remains the  
43 most dominant renewable energy fuel and its combustion maintains its position as the most  
44 prevalent and widespread technique for heating buildings [2].

45 Growing steadily for several years in France, renewable energy accounted for 12.9% of  
46 primary energy consumption and 19.1% of GFEC in 2020 [4,5]. Therefore, sustainable energy  
47 is considered the fourth major primary energy source following nuclear (40%), fossil fuels  
48 (28.1%), and natural gas (15.8%) [5]. Among all sustainable energy mixes, the wood-to-energy  
49 is still the leading renewable energy in France with a share of 32.9%, ahead of hydro (18.7%),  
50 wind (12.3%), biofuels (10.3%), aerothermal/geothermal/hydrothermal heat pumps (9.3%),  
51 renewable waste (4.3%) photovoltaic/solar (4.1), biogas (3.9%) and others (3%) including  
52 geothermal and tidal power [4,5].

53 Owing to the objectives of the European Union (EU) Directive 2009/28/EC to elevate  
54 the share of renewable energy to 32% by 2030 and 100% by 2050, the market for biomass  
55 boilers has known a very sharp rise in France these last years. According to the data released  
56 by the French Syndicate of Biomass Boilermakers, the first half of 2021 has recorded an  
57 increase of +150% and + 89% compared to the same half of 2019 and 2020, respectively. This  
58 sector is experiencing exceptional and sustained growth, boosted by the substitution of oil-fired  
59 appliances. Starting from 1 July 2022, public authorities will ban the potential installation of  
60 new oil-fired boilers in new construction and renovation projects. Besides, the entry into force  
61 of the environmental regulation RE 2020 has prohibited the installation of gas heating systems

62 in new individual housings. This is due to many factors including the high emissions of  
63 greenhouse gases, the high cost of natural gas (government prices soaring from 2021), and its  
64 importation from Norway, Russia and the Netherlands. In 2028, the objective is to reduce the  
65 climate impact of all residential buildings to 6.5 kg CO<sub>2</sub>eq/m<sup>2</sup>/year. Therefore, the building  
66 owners have to seek alternative cleaner and cost-effective equipment and fuels such as heat  
67 pumps (air-to-water heat pumps, geothermal heat pumps, etc.), district heating networks (50%  
68 of the primary energy from biomass or urban waste) or biomass combustion (wood boilers/  
69 stoves) [6].

70           Currently, forestry wood (traditional biomass) and agricultural residues are the cheapest  
71 energy on the French and international markets. These resources are abundant, spreadable over  
72 the French territory and easily stored in the form of pellets (modern biomass), which promotes  
73 local employment while making savings. For these reasons, the demand for biomass-based  
74 boilers/stoves consuming pellets, derived from wood or agricultural residues, as feedstock will  
75 be growing as being more efficient, economical and environmentally friendly than the other  
76 substitutes [6,7]. Firstly, the calorific value of 2 kg of pellets is equivalent to that of one liter of  
77 fuel oil and thus, a 3 m<sup>3</sup> pellets container corresponds to 1000 L of fuel oil. Besides, the high  
78 energy content of the pellets is solely converted into useful heat with an efficiency ranging from  
79 80 to 90%. Secondly, pellets are cost-effective, less expensive than oil and natural gas, and  
80 independent of the price fluctuations of these latter products on the market. Thirdly, the pellets  
81 revalorize wood residues without deforestation, consequently, they have a very low embodied  
82 energy, low environmental footprint, and a neutral carbon balance. This fits within the  
83 framework of the industrial decarbonization incentives of the French national recovery plan  
84 which strategy consists of the conversion of the existing fossil fuels/coal-based facilities into  
85 modern biomass-based ones that will be used at an industrial scale for heat production. Thus,

86 the combustion of pellets has the potential to emerge as a leader in the EU zero CO<sub>2</sub> emission  
87 strategy.

88 Pellets are a compact biomass fuel having distinctive advantages over raw biomass such  
89 as standardized characteristics, elevated calorific value, high density, and consequently low  
90 transportation, storage and handling fees [7]. In recent years, the pellet sector has grown  
91 spectacularly to become one of the most important and competitive biomass commodities  
92 manufactured in Europe (55%) and marketed internationally. At present, in some European  
93 countries such as France, Switzerland, Italy, Finland, Sweden, Austria, and Germany, the  
94 heating technology of biomass pellets is sufficiently mature to successfully compete with  
95 traditional oil and gas heating systems [2,3]. While an average gas-heated home emits about 5  
96 tons of CO<sub>2</sub> per year, the emissions of the same home heated with biomass (meeting the RE  
97 2020 standards) will be reduced to 0.5 tons of CO<sub>2</sub> per year, hence ten times less than the former  
98 [8]. Therefore, selecting a biomass pellet heating system is a profitable solution on the  
99 environmental, economic and energy levels.

100 In France, the pellet sector is looking forward to 2022 with confidence despite the  
101 uncertainties related to the supply of raw materials. According to the French norm (NF) and  
102 European Norm (EN) NF EN ISO 17225-1, virgin wood originates from untreated and  
103 unpainted Class A wood. This latter includes wood deriving from sawmills and wood industry  
104 by-products such as stems, logs, chips, sawdust, shavings, and chemically untreated woody  
105 materials [9]. After collection, the products are sorted, crushed, screened and mild dried prior  
106 to their compaction and pelletization at a temperature between 60 and 100 °C. Subsequently,  
107 the pellets are cooled and finely sieved to ensure the elimination of all dust that may obstruct  
108 or damage the combustion chamber. Before their packaging, the granules are tested to guarantee  
109 their quality before their commercialization. In France, pellet grades are governed by  
110 international norms (EN ISO 17 225-2) and European, German or French certifications namely

111 ENplus, DINplus, and NF Biocombustibles granules - biofuel pellets, respectively. These  
112 standards present quite similar requirements and differ notably in terms of heating value and  
113 density [10]. Thanks to their low moisture content (<10 wt.%) and high density (>600 kg/m<sup>3</sup>),  
114 pellets are considered high-energy performance fuels having the best combustion properties  
115 with an efficiency ranging from 85 (conventional boiler) to 105% (condensing boiler) based on  
116 fuel's net calorific value. However, the combustion of biomass pellets entails both particulate  
117 and gaseous emissions which can significantly impact air quality and, consequently, human  
118 health [11–15]. Numerous studies have emphasized the release of numerous pollutants such as  
119 nitrogen oxides (NO<sub>x</sub>) and sulfur oxides (SO<sub>x</sub>) [16–22], carbon monoxide (CO) [16–19,21–24],  
120 light unburned hydrocarbons (C<sub>x</sub>H<sub>y</sub>) [23], and particulate matter (PM) [11–13,21,22,25,26],  
121 from domestic boilers under standard laboratory conditions at full/nominal or reduced load  
122 operations [21,27]. Therefore, regulations on emissions are required to ensure that inhabitants  
123 are not exposed to toxic gases and dust and to incentivize manufacturers to improve their  
124 products to achieve the lowest possible emissions. For this reason, the labeling of the appliance  
125 and type of biomass is crucial. Several European countries have implemented quality labels or  
126 regulations including France which has established the “Flamme Verte” label for residential  
127 biomass heating systems in harmony with the NF EN 303-5 standard [28].

128 Today, pellets consumption (1.9 million tons) has slightly exceeded production (1.7  
129 million tons). Consequently, France has turned to the importation of around 0.4 million tons of  
130 pellets from Belgium, Spain or Germany to achieve the territory's self-sufficiency. Although  
131 the import remains limited (between 15 and 20%), french producers are searching for waste  
132 biomass to expand the spectrum of pellets in combustion systems. As the wood residues are not  
133 sufficient to meet the future growing demand for wood pellets, it is, therefore, necessary to find  
134 alternative biomass fuels to avoid unsustainable harvesting of forests as it will lead to adverse  
135 repercussions on the conservation of water, soil and biodiversity. Furthermore, it is essential to

136 diversify the supply of biomass to alleviate the pressure on noble wood. Several research papers  
137 investigated the use of various biomass residues in residential boilers. Verma et al. [29] studied  
138 two types of wheat straw, wood, peat, sunflower husk, pectin waste from citrus shells, reed  
139 canary grass, and apple pomace in a 40 kW domestic boiler. Carvalho et al. [30] evaluated the  
140 performance of a 15 kW pellet boiler operated with eight various agricultural fuels (wood with  
141 5% rye flour, Sorghum, hay, vineyard pruning, wheat bran, maize, Miscanthus and straw).  
142 Vineyard pruning and Miscanthus were the best fuels tested while straw and Sorghum have not  
143 satisfied the European legal requirement in terms of dust emissions. González et al. [31]  
144 investigated the burning of olive stone, forest pellets, cardoon, and tomato in a 12 kW boiler.  
145 Olive stones and tomato pellets were found to be excellent alternatives to wood pellets. In  
146 another paper, the authors investigated the burning of almond peel, almond shells and almond  
147 tree pruning. They identified that mixing this latter with 25% of wood pellets increased the  
148 combustion efficiency [32]. Miranda et al. [33] studied the combustion of forest wood and olive  
149 pomace pellets. The emissions from olive pomace were exceeding the EN 303-5 standard and  
150 therefore a blending using less than 50% of this product was recommended. Similarly, Limousy  
151 et al. [16] examined the burning of spent coffee grounds and pine sawdust. They noted that a  
152 50/50 mass proportion is needed to reach the NF standard requirements for agro-pellets.  
153 Likewise, several studies have investigated the blending of various biomass fuels such as  
154 woody, non-woody and herbaceous biomass, and evaluated their performance in small-scale  
155 combustion appliances [34–38]. These studies have highlighted that blending fuels with less  
156 problematic biomass (pure wood) seems to be efficient in enhancing the fuel properties,  
157 mitigating the challenges associated with their mineral composition) and achieving stable and  
158 high-performance operation of the boiler. Correspondingly, Zeng et al. [37] demonstrated that  
159 some binary blends including wood with different herbaceous biomasses (exhibiting critical  
160 element concentration) reduce substantially the gaseous and total particulate matter emissions.



161 Few studies have highlighted the energy recovery from Class B wood and construction  
162 and demolition (C&D) waste including the non-hazardous treated wood with a small  
163 concentration of chemical additives, for domestic heating [39–41]. The study conducted by  
164 Rabaçal et al. has revealed that the industrial wood residue wastes originating from wood  
165 products (C&D work residues, furniture and pallets) at their end-of-life (EoL) may be  
166 considered sustainable alternative fuels to be fired in domestic pellets boilers [39]. Another  
167 study has published that bioenergy may be produced from C&D wastes [40]. However, some  
168 toxic pollutants such as dioxins may be released during the combustion of C&D wood [41]. In  
169 light of this situation, biomass-based wastes have to be recovered at EoL as they may constitute  
170 a new potential viable energy resource for residential heating.

171 In this context, low chemically treated biomass-based insulation materials at EoL may  
172 be considered as alternative biomass resources to wood pellets. These non-dangerous wastes  
173 are currently neither reusable nor recyclable and are projected to end up dumped in landfills by  
174 2050, hence contributing to environmental risks [42]. To date, the treatment of bio-based  
175 insulation wastes is not addressed in the literature. It is, therefore, necessary to shed the light  
176 on the EoL of biomass-based insulation materials. Energy recovery via waste-to-energy  
177 conversion techniques was explored to be a promising strategy for the management of this type  
178 of waste [42]. For this reason, the combustion of these future wastes could provide an  
179 interesting supply solution from economic, energy and environmental points of view, boosting  
180 the sustainable economy, decarbonizing the heat and preserving the ecosystem by the release  
181 of biogenic CO<sub>2</sub>, respectively. Among all the biomass-based insulation materials described by  
182 Rabbat et al. [42], hemp fiber (HF) and wood fiber (WF) panels were selected as being the two  
183 low-chemically treated insulation materials with high assessed future deposits of 2,500 and  
184 40,000 tons in France by 2050, compared to the other materials. Besides, the fibrous nature of  
185 these materials facilitates their transformation into pellets at an industrial scale.

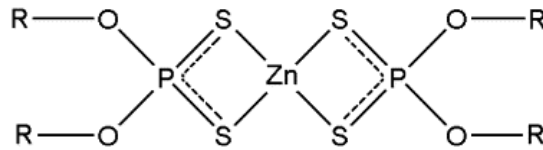
186 The main goal of this study is to evaluate the potential use of HF, WF and blends (HF and  
187 WF with certified wood pellets), as promising feedstock pellets in horizontal small-scale  
188 biomass boilers for domestic heating purposes. First, the fuels were pelletized and  
189 characterized. Then the performance of the boiler burning *DINplus* certified wood pellets  
190 currently commercialized in the French market was initially performed. Subsequently, the  
191 examination of the boiler efficiency and the characterization of the gaseous and particulate  
192 matter emissions of HF, WF and the blends, were realized in the same multi-fuel biomass boiler.  
193 Finally, the composition of the bottom ash was examined and its potential application was  
194 proposed.

## 195 2. Materials and methods

### 196 2.1. Pellets production and characterization

197 The *DINplus* certified wood pellets, HF and WF insulation panels were purchased from  
198 the French market. HF consists of 90% hemp fibers, <10% of synthetic fibers, and 0.2% of  
199 fungicide while WF is made from 95% of wood fibers, and 5% polyolefin fibers mixed with  
200 ammonium sulfate (flame retardant). Due to their heterogeneity and low bulk density, biomass-  
201 based insulation panels must be densified through pelletization. First, a grinder and refiner (T1  
202 of 5.5 kW power) manufactured in France by the TOY Group were used to reduce the  
203 granulometry of the products. Consequently, the shredded products were compacted and  
204 pelletized using an industrial granulated press unit (Type B360), manufactured by Promil Stolz  
205 in France. A high-performance semi-synthetic grease of type “CTDMEP 2” has been used for  
206 the lubrication of the metallic surfaces (highly-loaded bearings) of the pelletizer. This complex  
207 lithium grease is made of a base fluid (mineral oil lubricant with less than 3% dimethyl  
208 sulfoxide), a thickener so-called lithium complex soap (obtained from the reaction of fatty acids  
209 with lithium hydroxide monohydrate) [43], and performance additives (up to 2.5 wt.%) such as

210 zinc dialkyl-dithiophosphates (ZDDP) [44] (Fig. 1). Therefore, this grease may be found in  
 211 trace quantities in the pellets (< 2 to 3 wt.%).



213 *Fig. 1. Zinc dithiophosphate structure (R symbolizes the alkyl group [45])*

214 Next, the pellet fuels were characterized according to the relevant European and French  
 215 standards techniques (Table 1). The analyses were executed by the NF EN ISO/CEI 17025  
 216 certified laboratories of SOCOR (Environmental analysis laboratories located in Dechy,  
 217 France). Then, the characteristics of the wood-based pellets were compared to the certified  
 218 wood pellets following the NF EN ISO 17225-2 method (Graded wood pellets) and  
 219 commercialized on the French market to be incinerated in domestic heating boilers and stoves.  
 220 The hemp-based pellets were compared to the certified cereal straw pellets following the NF  
 221 EN ISO 17225-6 (Graded non-woody pellets) method and commercialized on the French  
 222 market to be incinerated in domestic heating boilers and stoves.

223 *Table 1. The tested properties realized on the pellets and their corresponding standards*

| <b>Pellets properties</b>                      | <b>Standard</b>    |
|--|--------------------|
| <b>Size</b>                                    | NF EN ISO 17829    |
| <b>Humidity</b>                                | NF EN ISO 18134-2  |
| <b>Mechanical durability</b>                   | NF EN ISO 17 831-1 |
| <b>Bulk density</b>                            | NF EN ISO 17828    |
| <b>Fine contents</b>                           | NF EN ISO 18846    |
| <b>Ash content</b>                             | NF EN ISO 18122    |
| <b>Calorific values</b>                        | NF EN ISO 18125    |
| <b>Carbon, hydrogen, and nitrogen contents</b> | NF EN ISO 16948    |
| <b>Sulfur and chlorine</b>                     | NF EN ISO 16994    |
| <b>Minor metals (Cd, Cr, Cu, Ni, Pb ...)</b>   | NF EN ISO 16968    |
| <b>Cellulose, hemicellulose, and lignin</b>    | FD U44-162         |
| <b>Ash fusibility</b>                          | NF EN ISO 21404    |

224  
 225 Noted that the ISO 17225 series of standards covers the requirement of using graded  
 226 pellet quality originating from wood and agricultural residues for public buildings, residential

227 and small commercial applications. These reasons that entail the use of high-grade fuel are  
228 typically due to the location of the small-scale installations in residential areas along with the  
229 lack of advanced controls on the operation and emissions. Besides, the agricultural pellets have  
230 high ash, low calcium (Ca) content, and high levels of some major elements such as chlorine  
231 (Cl), potassium (K), silicate (Si), nitrogen (N), and sulfur (S) content. When agricultural  
232 residues are intended for combustion, the risk of corrosion in small-scale boilers and gas venting  
233 systems should be considered. It is noteworthy that the contents of chlorine, phosphorus, and  
234 potassium elements in the straw may potentially generate chlorides, phosphates, and other  
235 chemical compounds that in turn may result in high emissions of chemically active ash with a  
236 low melting temperature triggering clogging, slag formation and corrosion in the boiler.

## 237 2.2. Test setup and boiler

238 The boiler used in this study is an HKRST-FSK-20 kW manufactured by the REKA industry  
239 in Denmark. This equipment is suitable for burning almost all types of fuels with up to 30% of  
240 moisture content such as wood pellets, straw, coal, wood chips, wood shavings, cereals, and  
241 cereal wastes. This high-effective steel plate boiler is fitted with a hopper in which the pellets  
242 are manually inserted and stored. The hopper is equipped with an impulse-controlled horizontal  
243 screw feeder that conveys the fuel onto a moving pushing grate to the combustion chamber  
244 where the total burning takes place. At first, ignition occurred manually. Dampers were  
245 manually opened allowing 5% primary and 100% secondary air to flow inside the appliance.  
246 The primary air is added under the grate and is conveyed through the openings of the grate to  
247 ember the ashes completely, helping in drying, volatilization and burnout of the char bed. The  
248 secondary air is injected into the flame zone to enhance the turbulence and minimize incomplete  
249 combustion [46]. The special feature of this boiler is its movable grates allowing it to accept  
250 multiple types of fuel as the movement of the grates prevents slag formation and the automatic  
251 transportation of the ash to their corresponding container ensuring efficient ash removal.

252           The schematic illustration of the small-scale pellet boiler installation is represented in  
253 Fig. 2. Several thermocouples were inserted to monitor and measure the temperature of the  
254 exhaust gases at different points of the system. These temperatures are essential for the  
255 determination of the overall boiler efficiency. A software was used to visualize and control the  
256 boiler operation as well as to instantaneously record the output gases, the burner power and the  
257 temperature values at different places.

258           Both heating and hot water could be provided by the boiler for commercial buildings or  
259 residential homes. In the present case, the boiler is connected only to an air heater. The hot  
260 combustion gases are used to heat the water inside the primary heat exchanger integrated within  
261 the boiler. This heat exchanger recovers the latent heat from steam and removes the condensed  
262 water from the boiler while maintaining the combustion. Then, the high-temperature water  
263 produced circulated via a network of insulated pipes (hoses) to feed an air heater consisting of  
264 a heat exchanger and a fan. When operating, this unit will be blowing hot air into the working  
265 area. The heat output to the water was obtained from the temperatures entering and exiting the  
266 air heater and the water flow rate which was measured at the inlet of the air-heater using a flow  
267 meter.

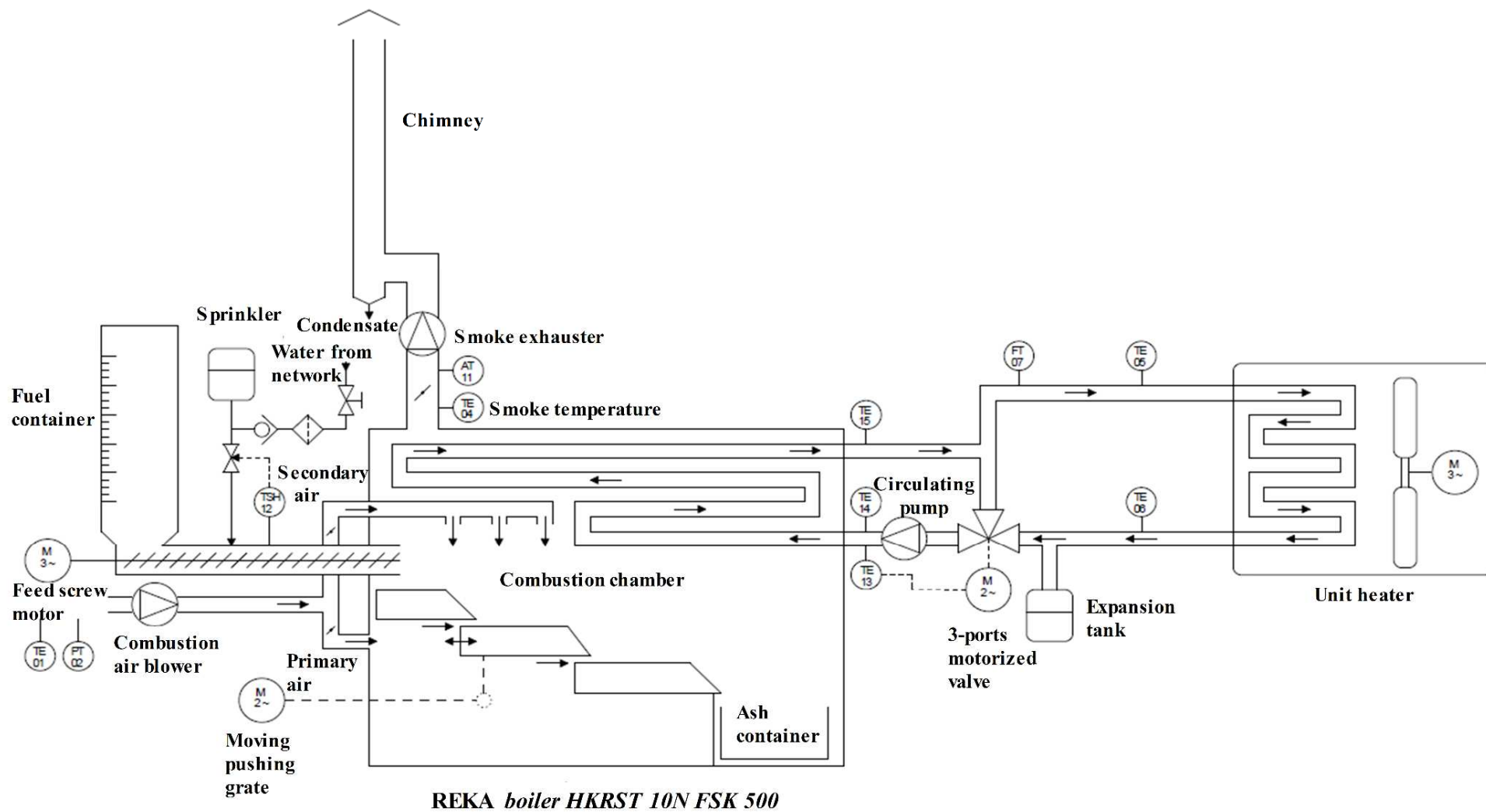


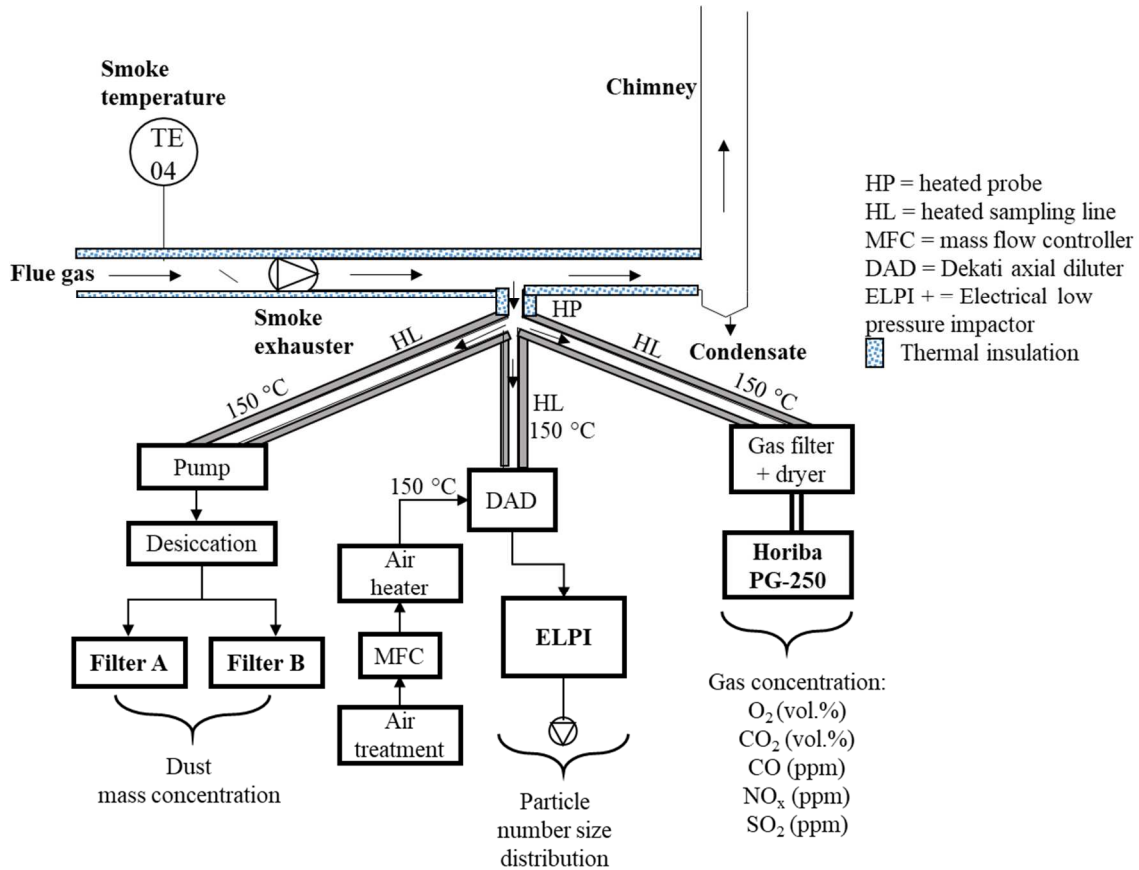
Fig. 2. The schematic diagram of the small-scale multi-fuel boiler

268

269

270 2.3. Emissions measurement

271 The schematic of the experimental setup for sampling and emissions measurement is illustrated  
 272 in Fig. 3.



273

274 *Fig. 3. The experimental setup for sampling: gaseous and particulate emissions measurement*

275 2.3.1. Particles examination

276 The sampling setup consists mainly of an Electrical Low-Pressure Impactor (ELPI)  
 277 manufactured by Dekati (Finland) and used to measure, in real-time, the size distributions and  
 278 the number concentrations of the PM dispersed in the flue gas exhausting from the boiler. This  
 279 analysis is preceded by a dilution system using filtered and heated air to prevent the automatic  
 280 saturation of the instrument. In this study, the set-up consisted of a two-step dilution in which  
 281 the exhaust gases are first heated to 150 °C (to eliminate the effects of condensation and  
 282 nucleation) before moving to ambient temperature. Based on the calibration curve provided in  
 283 the diluter datasheet, the dilution ratio at a pressure of 1013.3 mbar is 8.38. Therefore, the  
 284 combustion gas passes through an insulated connecting line heated at 150 °C before reaching

285 the ELPI. The apparatus through impaction, a physical mechanism, captures fine particles  
 286 having a diameter in the range of 30 nm to 10  $\mu\text{m}$ . The working principle of ELPI is separated  
 287 into particle charging, size classification by the cascade impactor, and electrical detection. First,  
 288 the particles are electrically charged in a positive unipolar particle charger and strike the 12-  
 289 stage impactor, which splits them into 12-size fractions based on their aerodynamic diameter  
 290 and inertia. Each stage is made up of two plates, including one with smaller orifices through  
 291 which particles can penetrate to perform a sharp turn (following the air streamlines) and thus  
 292 reach the subsequent lower plates. Particles whose aerodynamic diameter is greater than the  
 293 cut-off diameter of the stage will not be able to pass through the orifice due to its high inertia  
 294 and will therefore impact the second plate, “the collection plate”, and be deposited on it. The  
 295 cut-off diameters of the orifices, called nozzles, being smaller and smaller with each stage, it is  
 296 clear that the particles will be fractionated progressively, leading to the capture of nanoparticles  
 297 at the last stage. The high sensitivity electrometers count the charged aerosol particles by  
 298 measuring the total charge carried out by the particles on each plate. The particle's number is  
 299 calculated based on measured current using mathematical algorithms assuming a particle  
 300 density of  $1.9 \text{ g.cm}^{-3}$ . Then, particles are classified based on their cut-off aerodynamic diameters  
 301 (Table 2). The sampling was realized under isokinetic conditions as the gases were continuously  
 302 sampled by the ELPI at a constant suction flow rate of  $10 \text{ L.min}^{-1}$  while maintaining the same  
 303 speed as at the sampling point. The sampling duration was 20 minutes to avoid the failure of  
 304 the isokinetic conditions in case of high particle load and the measurements were repeated five  
 305 times for each combustion experiment. For each stage, the mean value of the collected particles  
 306 was calculated for 20 minutes and then used for calculating the overall mean value.

307 *Table 2. Cut-off diameters of the electrical low-pressure impactor*

| Stage                                      | Filter stage | 1      | 2      | 3     | 4      | 5      | 6      | 7      | 8      | 9      | 10     | 11     |
|--|--------------|--------|--------|-------|--------|--------|--------|--------|--------|--------|--------|--------|
| <b>Diameter (<math>\mu\text{m}</math>)</b> | 0.0131       | 0.0221 | 0.0415 | 0.072 | 0.1274 | 0.2095 | 0.3305 | 0.5316 | 0.8714 | 1.3968 | 2.2176 | 4.5352 |

308



309 2.3.2. Dust total concentration

310 The hot-filters sampling method was carried out to measure the mass concentration of  
311 total coarse and fine particles and soot (unburned carbon) formed during the combustion of the  
312 inlet feedstock. The sampling probe having a diameter of 2 cm was placed centrally in the flue  
313 gas cross-section. Four circular 47 mm porous glass microfiber filters were placed  
314 perpendicularly to the exhaust gas flow to reduce the collection of coarse particulates. The  
315 filters were pre-dried at 105 °C for 24 hours before being placed between a grid and a seal. The  
316 pump flow rate was regulated at 0.5 m<sup>3</sup>.h<sup>-1</sup>. After turning on the pump, the flue gas flows into  
317 the system and is sampled for a fixed period of 15 minutes where total particulates are collected.  
318 The weight of the filters was measured before and after the sampling using a balance with +/-  
319 0.1 mg precision. Consequently, the total mass concentration of the dust particles was deduced.  
320 Then, the mean value and standard deviation of the mass concentration of the particles collected  
321 on the four filters used were computed. The values are expressed in mg.Nm<sup>-3</sup> at 10 vol.% O<sub>2</sub>  
322 content in the dry flue gas. Moreover, X-ray Fluorescence spectroscopy was used to determine  
323 the type of inorganics and metals contained in the collected dust and bottom ash fractions.

324 2.3.3. Gaseous emissions

325 The concentration of the gaseous emissions released from the boiler was continuously  
326 measured using Horiba PG-250 portable gas analyzer in real-life conditions, once the operation  
327 of the boiler is stabilized. Emission values were recorded over approximately 3 hours and  
328 subsequently the overall mean and standard deviations were calculated for each gaseous specie  
329 analyzed. Before the measurement, the flue gas passes through an insulated heating probe to a  
330 gas filter and condensation dryer in which particulate matter and water vapor are removed,  
331 respectively. Next, the particulates-free dried gas enters the analyzer in which the instantaneous  
332 sampling was performed at a constant flow rate of 0.4 L.min<sup>-1</sup> (NF EN 304:2017 standard) and  
333 the continuous monitoring of the concentration of the five-dominant species (NO<sub>x</sub>, SO<sub>2</sub>, CO<sub>2</sub>,  
334 CO, and O<sub>2</sub>) is realized. This reliable and versatile analyzer uses the non-dispersive infra-red

335 (NDIR) absorption using an optical-*pneumatic* detector for CO and SO<sub>2</sub>; NDIR with a  
336 pyroelectric detector for CO<sub>2</sub>, Chemiluminescence detection method (CLD) by cross-flow  
337 modulation for NO<sub>x</sub> and Galvanic cell for O<sub>2</sub> monitoring. The instrument was calibrated with  
338 proper zeros and span gases.

339 • **Normalization of the gaseous emissions**

340 All the measurements were presented at 10 vol.% O<sub>2</sub> content in the dry flue gas to  
341 comply with environmental legislations. Eq. (1) was used to normalize the values of the  
342 emissions according to the referenced O<sub>2</sub> concentration in flue gases as excess air is supplied  
343 to ensure that a sufficient amount of O<sub>2</sub> is available for burning the fuel. The concentration of  
344 the gaseous emissions is therefore diluted by the excess air and needs to be corrected prior to  
345 being compared to local regulations.

$$[G_N] = [G] \times \frac{20.9 - O_2(ref)}{20.9 - O_2} \quad (1)$$

346 Where [G<sub>N</sub>] and [G] are the normalized and measured gas concentration  
347 O<sub>2</sub> (ref) and O<sub>2</sub> are the reference and measured oxygen percentages, respectively (vol.%)

348 • **Excess air**

349 The deficiency in burning air reduces fuel efficiency, generates soot, and forms highly  
350 toxic gases such as CO. To be certain that an adequate amount of oxygen is available to  
351 completely react with the feedstock and ensure sufficient mixing, an oxygen surplus is usually  
352 needed and known as Excess air. This latter represents the percentage of air (non-reacted O<sub>2</sub>)  
353 exceeding the theoretical amount required for complete combustion and is expressed as follows:

$$Excess\ air\ (\%) = \frac{O_2}{20.9 - O_2} \times 100 \quad (2)$$

354 Where O<sub>2</sub> is the measured oxygen percentage (vol.%)

355 Another expression that could be used to describe the excess air is Lambda (λ) as follows:

$$\lambda = \frac{\text{Excess air (\%)}}{100} + 1 \quad (3)$$

356 The excess air,  $\lambda$ , is the most important parameter to control fuel consumption, boiler efficiency  
 357 and gaseous emissions.

358 • **CO<sub>2</sub> concentration**

359 The volumetric concentration of CO<sub>2</sub> in the exhaust gas (vol.%) was calculated as the following:

$$CO_2 = CO_{2_{max}} \times \frac{20.9 - O_2}{20.9} \quad (4)$$

360 Where O<sub>2</sub> is the concentration of oxygen in the combustion gases (vol.%)

361  $CO_{2_{max}}$  is the maximum concentration of CO<sub>2</sub> (vol.%).

362 This latter term is calculated based on the fuel stoichiometric combustion under theoretical air  
 363 and elemental analysis. This parameter refers to the number of CO<sub>2</sub> molecules generated by the  
 364 complete combustion of the fuel divided by the total number of molecules of combustion  
 365 products. For wood fuel, the  $CO_{2_{max}}$  in dry gases is around 19.7% by volume [47]. This value  
 366 can be readily calculated for biomass fuel's dry gases as follows:

$$CO_{2_{max}} (\%) = \frac{[C]}{[C] + \frac{79.1}{20.9} \times \left( [C] + \frac{[H]}{4} - \frac{[O]}{2} \right)} \quad (5)$$

367 Where [C], [H] and [O] represent the molar fraction of carbon, hydrogen and oxygen of each  
 368 biomass feedstock (mol.%), respectively.

369 2.4. Boiler performance

370 To assess the boiler performance, the boiler efficiency should be computed. This could  
 371 be performed via two possible methods: an indirect or a direct method. The former method  
 372 qualifies and quantifies all the losses that may occur in the boiler (convection/irradiation losses,

373 on-off losses, blowdown losses, etc.) whereas the latter method is an energy balance based on  
374 the ratio between the heat recovered and the heat supplied by the fuel [48–50]. During the  
375 efficiency computation, the boiler’s electricity consumption was not considered.

#### 376 2.4.1. Indirect boiler efficiency (heat loss method)

377 The indirect boiler efficiency (IBE) measures the ability of a burner to burn fuel while  
378 considering all heat losses. Depending on the magnitude of each computed loss, some problem  
379 areas may be observed and hence corrective actions (optimizing excess air, minimizing the  
380 stack losses, and reducing fuel consumption) may be implemented for rectification. IBE is  
381 calculated according to DIN 1942 method and British Standard BS845: Part 1: 1987 by  
382 subtracting from 100% the individual heat losses ( $L_i$ ) using the following expression [48–50]:

$$IBE(\%) = 100 - HL_i \quad (6)$$

383 where  $HL_i$  stands for individual losses (%)

384 While many other losses may be unavoidable as these owing to moisture in the air,  
385 unburned carbon in ash, convection and irradiation, the flue gas losses (FGL) are the foremost  
386 and largest source of heat loss and energy wasted in boilers. These losses rely on the  
387 composition of the fuel, the combustion conditions and the temperature of the discharged gas.  
388 If the burner is properly maintained, the other losses are minimal and could be neglected. FGL  
389 losses are the summation of the sensible heat energy in dry flue gas ( $HL_1$ ), enthalpy of water  
390 vapor’s latent heat produced due to hydrogen in the fuel ( $HL_2$ ), the heat loss owing to the  
391 moisture content in the feedstock ( $HL_3$ ) and unburned fuel in the flue gas ( $HL_4$ ) mechanisms;  
392 calculated using Eq. (7). The former losses occur when the exhaust gas has a temperature  
393 exceeding the ambient one and leaves the boiler via the stack, leading to the waste of a huge  
394 amount of energy. However, the latent heat energy originates from the presence of steam due  
395 to water production (from biomass moisture and hydrogen contents) and its evaporation from

396 the high-temperature flue gas. The latter represents the heat lost by incomplete combustion of  
397 the feedstock; based on the percentage of unburned gases (CO and C<sub>x</sub>H<sub>y</sub>).

$$FGL = HL_1 + HL_2 + HL_3 + HL_4 \quad (7)$$

398 • **HL<sub>1</sub>: Loss due to sensible heat in the dried exhaust gas**

399 This loss term measures the quantity of hot exhaust gases passing out of the chimney. This  
400 is one of the main losses in the boiler and is determined using the following expression [48,50]:

$$HL_1 = \frac{m_{FG} \times C_{p,FG} \times (T_f - T_a)}{HHV_{fuel}} \times 100 \quad (8)$$

401 Where  $m_{FG}$  is the summation of the mass of CO<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub> (excess), CO, NO<sub>x</sub> and SO<sub>2</sub> included  
402 in the dry flue gas (FG) without considering the water (kg dry FG/kg fuel)

403  $C_{p,FG}$  is the specific heat of the dried flue gas (kJ/kg °C)

404  $T_f$  and  $T_a$  are the temperature of the stack gases and ambient air, respectively (°C)

405  $HHV_{fuel}$  is the higher heating value of the fuel (kJ.kg<sup>-1</sup>)

406 • **HL<sub>2</sub>: Loss due to enthalpy of water vapor owing to hydrogen in the fuel**

407 This loss term (Eq. (9)) consists of the sensible heat to heat moisture to the boiling point,  
408 the heat of vaporization (latent heat) of water in the flue gases and the energy needed to heat  
409 the water vapor to exhaust temperature. During combustion, water is formed in a liquid state  
410 owing to the hydrogen content of the feedstock. This water is subjected to high temperatures in  
411 the combustion chamber where it is transformed into vapor (steam) carrying latent heat. This  
412 phase change absorbs a tremendous quantity of energy that is not recovered. When exiting the  
413 stack, the water vapor is condensed and its latent heat is thus provided to the atmosphere hence  
414 forming a white cloud on a cold day [48,50].

$$HL_2 = \frac{9 H \times [L_{v,S} + C_{p,SS} \times (T_f - T_a)]}{HHV_{fuel}} \quad (9)$$

415 Where H is the hydrogen content of the biomass feedstock (wt.%)

416  $C_{p,SS}$  is the superheated steam (SS) specific heat ( $C_{p,SS}=2.0267$  kJ/kg °C)

417  $L_{v,S}$  is the specific enthalpy of steam (total heat: latent and sensible) for partial pressure

418 ( $L_{v,S}=2675.43$  kJ.kg<sup>-1</sup>)

419 • **HL3: Loss due to enthalpy of water vapor owing to moisture in the fuel**

420 Similarly, all fuels entering the boiler contain moisture. This latter leaves as superheated  
 421 steam. The humidity loss consists of the sensible heat loss to heat the moisture to its boiling  
 422 point for ensuring its evaporation and the superheating of the resulting steam to reach the  
 423 exhaust gas temperature. This loss term may be calculated using this formula [48,50]:

$$HL_3 = \frac{MC \times [L_{v,S} + C_{p,SS} \times (T_f - T_a)]}{HHV_{fuel}} \quad (10)$$

424 Where MC is the moisture content of the biomass (wt.%)

425 • **HL4: Loss due to partial combustion products in the exhaust gas**

426 This loss term indicates the energy loss due to incomplete burning of the fuel and the  
 427 emissions of unburned gases including CO and unburned hydrocarbons (C<sub>x</sub>H<sub>y</sub>). The carbon  
 428 content of the fuel reacts first with O<sub>2</sub> to produce CO that in turn will be oxidized into CO<sub>2</sub>. As  
 429 the carbon is partially converted to CO owing to a lack of O<sub>2</sub> or poor mixing of fuel/air, energy  
 430 is lost. In this study, C<sub>x</sub>H<sub>y</sub> were not measured. Thus, this heat loss only depends on CO and is  
 431 expressed as follows [48,50]:

$$HL_4 = \frac{24033 \times C}{HHV_{fuel}} \times \frac{[CO]}{[CO_2] + [CO]} \quad (11)$$

432 Where 24033 is the heat loss due to partial combustion of carbon (kJ/kg)

433 C is the carbon content of the feedstock biomass (wt.%)

434 [CO] and [CO<sub>2</sub>] are the concentrations of CO and CO<sub>2</sub> in the flue gas, respectively (vol.%)

#### 435 2.4.2. Direct Boiler Efficiency (input-output method)

436 Direct boiler efficiency (DBE) relies on the proper fuel burning, efficiency of heat transfer,  
437 operation and maintenance. This efficiency could be directly computed based on NF EN 303-5  
438 standard by dividing the output energy (heat utilized) by the input energy (heat supplied) of the  
439 boiler as described hereafter [48–50]:

$$DBE = \frac{\text{Heat output}}{\text{Heat input}} \times 100 = \frac{P_{out}}{P_{in}} \times 100 \quad (12)$$

440 Where  $P_{out}$  is the nominal useful power energy of the burner (kW)

441  $P_{in}$  is the calorific mass flow (kW)

##### 442 • Heat output

443 The heat output (kW) is the energy used effectively to generate hot water, calculated as follows:

$$P_{out} = Q_w \times C_{Pw} \times (T_{in} - T_{out}) \quad (13)$$

444 Where  $Q_w$  is the mass flow rate of water entering the air heater unit (kg.s<sup>-1</sup>)

445  $C_{Pw}$  is the calorific capacity of water (kJ/kg °C)

446  $T_{in}$  and  $T_{out}$  are the temperatures of water entering and exiting the air heater, respectively (°C)

##### 447 • Heat input

448 The heat input (kW) is the energy provided by the gross calorific value of the burnt fuel,  
449 expressed by:

$$P_{in} = Q_{fuel} \times HHV \quad (14)$$

450 Where  $Q_{fuel}$  is the mass flow rate of the biomass pellets entering the boiler (kg.s<sup>-1</sup>)

451 HHV is the higher heating value of the fuel ( $\text{kJ}\cdot\text{kg}^{-1}$ )

## 452 3. Results and discussion

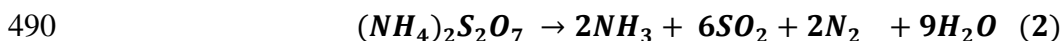
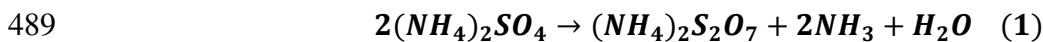
### 453 3.1. Quality of the pellets

454 In Europe, numerous standards (EN 14961-2, ...) and labels (NF, Swan, *ENplus*,  
455 *DINplus*...) are present for defining the minimum biomass pellet quality requirement for  
456 industrial and residential applications. In France, the NF EN ISO 17225-2 and NF EN ISO  
457 17225-6 officially state the characteristics of the wood and agro-based pellets to be  
458 commercialized on the French market, respectively. The standards specify an optimal diameter  
459 of 6 mm for efficient transportation of the pellets [21]. The physico-chemical properties of the  
460 different pellets as well as their chemical composition are summarized in Table 3.

461 As the *DINplus* certified wood pellets fall within the EN, they were taken as a reference  
462 for this study. Besides, the characteristics of the hemp-based pellets are following the NF EN  
463 ISO 17225-6 regulation for graded non-woody (straw) pellets, hence they can be marketed on  
464 the French market as new biomass fuel for boilers. However, some parameters of the wood-  
465 based pellets are not aligned with the NF EN ISO 17225-2 norm: the mechanical stability, the  
466 contents of fine particles, nitrogen and sulfur elements. Mechanical durability is an extremely  
467 relevant aspect to be considered for pellets as it describes the capacity of densified pellets to  
468 persist undamaged after shock or friction application. The wood-based pellets have a slightly  
469 lower mechanical durability than the standard (94.6 wt.% wb and 96.5 wt.% wb, respectively),  
470 hence fine particles may be generated when transporting and handling (loading or unloading)  
471 the pellets. This is attributed to the incorporated additives inside this feedstock that limit the  
472 natural binding of the wood particles. Another reason that could be at the source of this lower  
473 durability may be the improper cooling process of the pellets that induces cracks in the pellets  
474 owing to the stress between the warm center of the pellet and the cooled surface layer [51]. The



475 fine content (4 wt.% wb) is 4 times greater than the standard requirement ( $\leq 1$ wt.% wb). This  
 476 result is expected as the pellets were not sieved after the pelletization process. The fine content  
 477 might therefore comply with the standard if the pellets were sieved. The total nitrogen content  
 478 (1.08 wt.% db) is slightly higher than the standard ( $\leq 1$  wt.% db) whereas the sulfur content  
 479 (1.18 wt.% db) is significantly exceeding the values specified by the norm ( $\leq 0.05$  wt.% db).  
 480 This is attributed to the ammonium sulfate  $(\text{NH}_4)_2\text{SO}_4$ , an inorganic salt added to the wood  
 481 panels to improve their fire properties. This flame retardant starts melting at  $235^\circ\text{C}$  and  
 482 decomposes through the generation of non-flammable toxic gases ( $\text{SO}_2$  and  $\text{NH}_3$ ) in the flaming  
 483 zone [52]. The decomposition takes place via two distinct sets of endothermic reactions (1) and  
 484 (2). In the former step, the ammonium sulfate is fragmented into ammonium pyrosulfate,  
 485  $(\text{NH}_4)_2\text{S}_2\text{O}_7$  [53] that in turn will be broken down in the second step (dominant reaction) into  
 486  $\text{NH}_3$ ,  $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{N}_2$  at temperatures exceeding  $280^\circ\text{C}$  [54]. Under fire conditions, the  
 487 nitrogen atom of the ammonia molecule produces an elevated concentration of  $\text{NO}_x$  along with  
 488 nitrous oxide [55].



491 However, it should be noted that the sulfur present in the wood-based pellets is not  
 492 bounded to the organic fraction, hence sulfur will be mostly concentrated in the ash fraction  
 493 with some emissions of  $\text{SO}_2$ . As the ultimate analysis of the wood-based pellets does not fulfill  
 494 all the requirements of the NF Biocombustibles granules, particularly in terms of sulfur and  
 495 nitrogen contents, a blend of these pellets with certified wood is crucial to respect the standard.

496 *Table 3. Characteristics of the biomass pellets and quality requirements for pellet fuel based on standards for residential*  
 497 *application*

| Analytical parameters | Units | Experimental pellets | NF EN ISO 17225-2 | NF EN ISO 17225-6 |
|-----------------------|-------|----------------------|-------------------|-------------------|
|-----------------------|-------|----------------------|-------------------|-------------------|

|   |                           | <b>DIN<sub>plus</sub><br/>wood<br/>(WR)</b> | <b>Wood-<br/>Fiber<br/>(WF)</b> | <b>Hemp-<br/>fiber<br/>(HF)</b> | <b>Graded<br/>wood<br/>pellets</b> | <b>Graded<br/>non-<br/>woody<br/>pellets</b> |
|---|---------------------------|---|---------------------------------|---------------------------------|------------------------------------|--|
| <b>Analysis of the product</b>                                    |                           |   |                                 |                                 |                                    |  |
| <b>Size of the granules</b>                                       |                           |   |                                 |                                 |                                    |  |
| <b>Pellets less than<br/>3.15 mm</b>                              | wt.% wb                   | 0.0   | 1.3                             | 0.0                             | 0.0                                |  |
| <b>Granules between<br/>3.15 mm and 40<br/>mm</b>                 | wt.% wb                   | 100.0                                       | 98.7                            | 100.0                           | ≥99.0                              | ≥ 99.0                                       |
| <b>Pellets between 40<br/>mm and 45 mm</b>                        | wt.% wb                   | 0.0   | 0.0                             | 0.0                             | ≤1.0                               | ≤ 1.0  |
| <b>Granules larger<br/>than 45 mm</b>                             | wt.% wb                   | 0.0   | 0.0                             | 0.0                             | 0.0                                | 0.0  |
| <b>Average diameter</b>   | mm                        | 6.0   | 6.0                             | 6.0                             | 6.0                                | 6.0 to 10.0                                  |
| <b>Physical analysis</b>  |                           |   |                                 |                                 |                                    |  |
| <b>Preparation/grinding of a sample</b>                           |                           |   |                                 |                                 |                                    |  |
| <b>Total moisture</b>   | wt.% ar                   | 8.0   | 1.6                             | 2.2                             | ≤ 10                               | ≤ 10   |
| <b>Dry matter</b>   | wt.% wb                   | 92.0  | 98.4                            | 97.8                            |                                    |  |
| <b>Mechanical<br/>durability</b>                                  | wt.% ar                   | ≥ 97.5                                      | <b>94.6</b>                     | 99.2                            | ≥ 96.5                             | ≥ 97.5                                       |
| <b>Bulk density</b>   | kg.m <sup>-3</sup> ar     | ≥ 600                                       | 610                             | 740                             | ≥ 600                              | ≥ 600  |
| <b>Fine &lt; 3.15 mm</b>  | wt.% wb                   | ≤ 1   | <b>4.0</b>                      | 0.4                             | ≤ 1                                | ≤ 1  |
| <b>Additives</b>  | wt.% wb                   | 0   | ≤ 2                             | ≤ 3                             | ≤ 2                                | ≤ 5  |
| <b>Basic analysis</b>   |                           |   |                                 |                                 |                                    |  |
| <b>Ash at 550°C</b>   | wt.% db                   | ≤ 0.7                                       | 0.40                            | 2.57                            | ≤ 2.0                              | ≤ 6.0  |
|   | wt.% wb                   |   | 0.40                            | 2.52                            |                                    |  |
| <b>Elemental analysis</b>   |                           |   |                                 |                                 |                                    |  |
| <b>Mineralization for determination of halogens and/or sulfur</b> |                           |   |                                 |                                 |                                    |  |
| <b>Total carbon</b>   |                           | 49.62                                       | 49.4                            | 46.3                            |                                    |  |
| <b>Total hydrogen</b>   |                           | 6.06  | 6.66                            | 6.16                            |                                    |  |
| <b>Total nitrogen</b>   | wt.% db                   | ≤ 0.3                                       | <b>1.08</b>                     | 0.61                            | ≤ 1.0                              | ≤ 0.7  |
| <b>Total sulfur</b>   |                           | ≤ 0.03                                      | <b>1.180</b>                    | 0.044                           | ≤ 0.05                             | ≤ 0.1  |
| <b>Total chlorine</b>   |                           | ≤ 0.02                                      | 0.008                           | 0.09                            | ≤ 0.03                             | ≤ 0.1  |
| <b>Thermal analysis</b>   |                           |   |                                 |                                 |                                    |  |
| <b>Gross calorific<br/>value (HHV)</b>                            | MJ.kg <sup>-1</sup><br>db | 19.81                                       | 20.420                          | 18.167                          | ≥ 16.5                             | ≥ 14.5                                       |
| <b>Lower calorific<br/>value (LHV)</b>                            | MJ.kg <sup>-1</sup><br>db | 18.578                                      | 19.050                          | 16.898                          |                                    |  |
| <b>Metals</b>   |                           |   |                                 |                                 |                                    |  |
| <b>Mineralization for the determination of minor metals</b>       |                           |   |                                 |                                 |                                    |  |
| <b>Total cadmium</b>  |                           | ≤ 0.5                                       | < 0.4                           | < 0.4                           | ≤ 0.5                              | ≤ 0.5  |
| <b>Total chromium</b>   |                           | ≤ 10  | < 1                             | < 1                             | ≤ 10                               | ≤ 50   |
| <b>Total copper</b>   |                           | ≤ 10  | < 1.3                           | 2.5                             | ≤ 10                               | ≤ 20   |
| <b>Total Nickel</b>   | mg.kg <sup>-1</sup>       | ≤ 10  | < 1                             | < 1                             | ≤ 10                               | ≤ 10   |
| <b>Total lead</b>   | db                        | ≤ 10  | < 1                             | < 1                             | ≤ 10                               | ≤ 10   |
| <b>Total Zinc</b>   |                           | ≤ 100                                       | 12.4                            | 17.4                            | ≤ 100                              | ≤ 100  |
| <b>Total Arsenic</b>  |                           | ≤ 1   | < 1                             | < 1                             | ≤ 1                                | ≤ 1  |
| <b>Total mercury</b>  |                           | ≤ 0.1                                       | < 0.1                           | < 0.1                           | ≤ 0.1                              | ≤ 0.1  |

| <b>Ash fusibility</b>  |    |        |        |        |                                     |
|--|----|--------|--------|--------|-------------------------------------|
| <b>Fusibility of ash in an oxidizing atmosphere at 550°C</b> |    |        |        |        |                                     |
| <b>Initial shrinkage temperature</b>                         | °C | > 1000 | 820    | 640    | To be mentioned if exceeding 550 °C |
| <b>Deformation temperature</b>                               | °C | > 1200 | 1260   | 1160   |                                     |
| <b>Hemisphere temperature</b>                                | °C | > 1500 | > 1500 | > 1500 |                                     |
| <b>Flow temperature</b>                                      | °C | > 1500 | > 1500 | > 1500 |                                     |

498 ar: as received, db: on a dry basis, and wb: on a wet basis

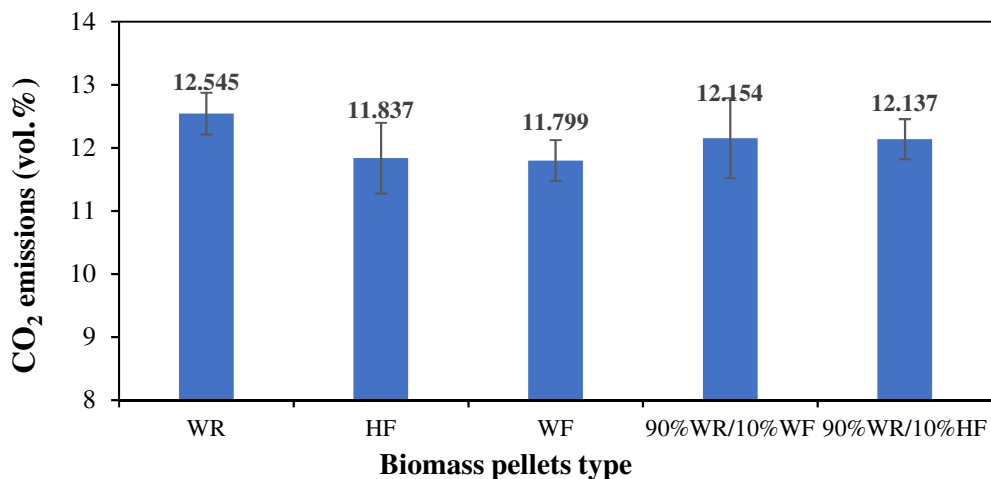
499 3.2. Gaseous emissions

500 In ideal-complete combustion, the flue gas exiting the boiler is only composed of CO<sub>2</sub>, N<sub>2</sub>  
501 and H<sub>2</sub>O. Nevertheless, in actual practices, the perfect combustion of biomass fuels barely takes  
502 place due to several factors such as the various pollutants contained within these feedstocks,  
503 the high combustion temperature, the low turbulence to blend the fuel with the required amount  
504 of oxygen and proper residence time to enable the biomass constituents to be consumed entirely  
505 [29,31,56,57]. Therefore, NO<sub>x</sub>, SO<sub>x</sub>, CO and C<sub>x</sub>H<sub>y</sub> are detected in the gaseous emissions and  
506 their concentrations are highly influenced by the fuel burnt. The real-life emission profiles of  
507 this biomass boiler operating in pulsed mode during the combustion experiment are displayed  
508 for each burnt fuel in the Supplementary material. The gaseous emissions were compared to the  
509 EN 303-5:2021 emission thresholds relative to automatically stoked heating boilers (<500 kW)  
510 for solid fuels.

511 Repeated measure ANOVA (analysis of variance) was carried out on Minitab to compare  
512 the effect of pellets type and operation time on the mean concentrations of the gaseous and PM  
513 emissions during combustion. When mean gas emissions were significantly different between  
514 pellets or during the combustion duration (p-value<0.05), a Bonferroni post-hoc test was  
515 performed to identify which pair means differed. The results of the ANOVA and Bonferroni  
516 pairwise comparison are presented in the Supplementary material.

517 3.2.1. Carbon dioxide emissions

518 During combustion, the CO<sub>2</sub> emissions (Fig. 4) are dependent on the C content of the fuel.  
519 The highest the C content, the highest the CO<sub>2</sub> production. Even if the results observed are very  
520 close, it is possible to identify a trend: WR has the higher C content (49.62 wt.%), its CO<sub>2</sub>  
521 emissions were the highest (12.545 vol.%). A lower concentration of CO<sub>2</sub> was related to hemp-  
522 based pellets as comprising the lowest C content (46.3 wt.%). Although its high C content  
523 (49.40 wt.%), the CO<sub>2</sub> concentration was the lowest for WF (11.799 vol.%) owing to the flame  
524 retardant treatment that interferes to suppress the decomposition of the pellets by the release of  
525 non-flammable gases. The CO<sub>2</sub> percentage increased in the blends.



526

527

Fig. 4. CO<sub>2</sub> emissions in vol.% of the different fuels and blends tested

528 3.2.2. Carbon monoxide emissions

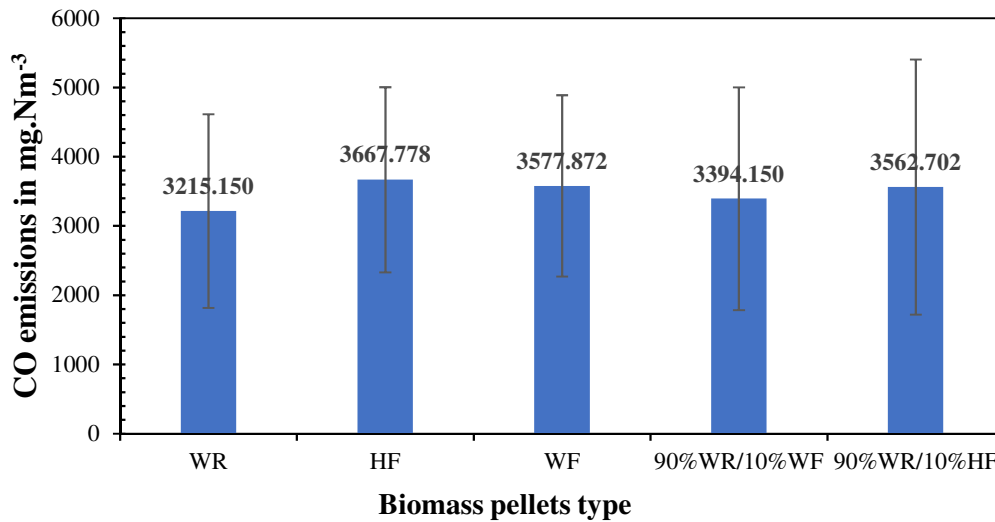
529 The concentrations of CO were high in the various cases studied (Fig. 5). However, the  
530 measured CO levels are in agreement with horizontal feed boilers, tested under standard  
531 laboratory conditions [29]. This result could be attributed to one or more of the cited reasons:  
532 the insufficient amount of O<sub>2</sub> (poor excess air), low burning temperature, low mixing of the  
533 feedstock with air or too short residence time of the gases in the burning chamber [21]. An  
534 excess air value between 1.2 and 1.5 was reported to be optimal to increase combustion  
535 efficiency and reduce heat losses [41]. Generally, a higher excess air lowers CO concentration.

536 In this study, the excess air value was around 1.6 for all the tested fuels. Nevertheless, the air  
537 and fuel are not properly mixed in the boiler, thus the continuous supply of excess air may have  
538 a reverse action by shortening the contact time between the biomass and O<sub>2</sub> and inhibiting the  
539 complete burnout; hence leading to a high concentration of CO and C<sub>x</sub>H<sub>y</sub>.

540 For all tested pellets, the CO concentration ranged between 3200 and 3670 mg.Nm<sup>-3</sup> with a  
541 standard deviation of 1000 mg.Nm<sup>-3</sup>. The high fluctuation of these values during the acquisition  
542 time is responsible for this high standard deviation, thus no significant difference can be noticed  
543 between the pellets. The highly intermittent nature of CO emissions, in this type of pulsed mode  
544 combustion, explains the larger standard deviations observed for this pollutant. Indeed, several  
545 studies have noticed a high standard deviation for CO emissions of the same order of magnitude  
546 varying between 30% and 50% of the mean value [53,54]. In this study, the standard deviation  
547 for CO was around 20 to 25% of the mean value for all the burnt fuels.

548 The DIN*plus* certified wood pellets have the lowest bulk density, hence they provide, for a  
549 similar flow rate, less power and higher excess air into the burning zone. Therefore, their CO  
550 emissions were comparatively low. The emissions of CO were slightly higher for HF and WF  
551 pellets. The discrepancy in CO emissions between agricultural and woody materials may be  
552 related to the furnace control system, i.e., the furnace was not capable of adjusting the burning  
553 air to the requirements of the various fuels. The combustion chamber of the multi-fuel boiler  
554 used in this study might not be set to receive chemically treated woody and non-woody fuels.  
555 The combustion of the flame-retardants and plastic binders incorporated in HF and WF could  
556 be also behind the higher CO emissions. These additives could have led to less suitable burning  
557 conditions. Another reason could be the slag formed in the bottom ash and accumulated during  
558 HF combustion in the grate of the burner. Ash agglomerates could have induced an irregular  
559 flow of burning air across the feedstock bed, which might, in turn, impede the burnout  
560 (oxidation) of the gas phase and char; hence increasing CO emissions and reducing the boiler

561 efficiency [11,30]. Although the boiler is equipped with moving grates, their motion should be  
562 operated more frequently for hemp fiber material to avoid the agglomeration and accumulation  
563 of ash [37].



564

565 *Fig. 5. CO emissions in mg.Nm<sup>-3</sup> of the different fuels. The error bars correspond to the standard error of the mean.*

566 For boiler (< 50 kW) classes 3, 4 and 5, CO emissions legal requirements in the EN 303-  
567 5:2021 are 3000, 1000 and 500 mg.m<sup>-3</sup> at 10% O<sub>2</sub>, respectively. The tested fuels can only fulfill  
568 the CO emissions permissible values for boiler class 3 while considering the standard deviation.

### 569 3.2.3. Nitrogen oxides emissions

570 In boilers, the burning of biomass contributes to the liberation of NO<sub>x</sub> (NO and NO<sub>2</sub>) and  
571 nitrous oxide (N<sub>2</sub>O). NO<sub>x</sub> lead to acid rains and atmospheric ozone formation that in turn are  
572 harmful to vegetation and cause eyes and lung irritations [58,59]. Throughout the burning, NO  
573 is primarily formed. However, this compound is unstable and hence undergoes an oxidation  
574 reaction and is converted into NO<sub>2</sub> once discharged into the atmosphere.

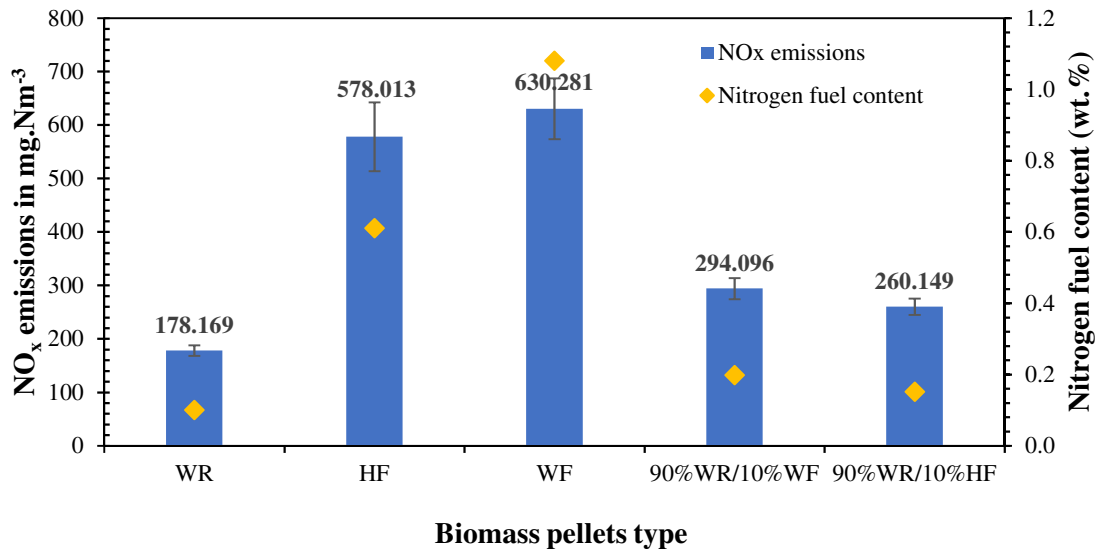
575 NO<sub>x</sub> emissions are usually linked to three major pathways: fuel-NO<sub>x</sub>, prompt NO<sub>x</sub> and  
576 thermal NO<sub>x</sub>. The former designates the NO<sub>x</sub> formed because of the oxidation reaction  
577 occurring between the oxygen from the air and the nitrogen chemically bound to the fuel.  
578 Prompt NO<sub>x</sub> is generated in the flame front (> 1300 °C) owing to the reaction between the

579 atmospheric N<sub>2</sub> and hydrocarbon radicals (CH<sup>•</sup>) that leads to the formation of hydrogen cyanide  
580 (HCN) that in turn will be oxidized in the flame zone to produce flame-generated NO<sub>x</sub>. Thermal  
581 NO<sub>x</sub> results from oxidation reactions occurring at very high temperatures (> 1300 °C) between  
582 atmospheric N<sub>2</sub> and oxygen or OH radicals [60]. In this study, the temperature of the flame  
583 inside the small-scale biomass boiler was calculated. The flame temperature fluctuated between  
584 1390 °C for HF and 1500 °C for WR. Therefore, NO<sub>x</sub> emissions are dominated by fuel-NO<sub>x</sub>  
585 with a small concentration originating from thermal NO<sub>x</sub> and prompt NO<sub>x</sub>.

586 In Fig. 6, NO<sub>x</sub> emissions were observed to be higher for wood fiber (630 mg.Nm<sup>-3</sup>) and  
587 hemp fibers (578 mg.Nm<sup>-3</sup>) than for DIN*plus* certified wood (178 mg.Nm<sup>-3</sup>). This is attributed  
588 to their greater N content (1.08 and 0.61 wt.%, respectively) than high-graded wood (< 0.3  
589 wt.%). However, nitrogen content and NO<sub>x</sub> formation are not linearly correlated as their  
590 emissions are influenced by several factors including the tendency of a material to form ash  
591 agglomerated and the excess air ratio [37]. The high fraction of NO<sub>x</sub> emitted by the combustion  
592 of HF could be due to the high N content as well as the high ash content of this material  
593 catalyzing NO<sub>x</sub> formation. Besides, when the fuel-N exceeds 1.1 wt.%, the combustion of the  
594 pellets leads to the formation of nitrogen-containing species, and the production of NO is  
595 reduced [61]. This can be due to the catalytic effect of ash surface, particularly when burning  
596 fuels with important ash contents and comprising active elements such as potassium, iron,  
597 magnesium and calcium [30,62]. N<sub>2</sub> could be formed from the reduction of fuel-N when  
598 performing understoichiometric primary combustion followed by secondary combustion [30].  
599 Concerning legal requirements, the NO<sub>x</sub> emissions threshold is stipulated to be 200 mg.Nm<sup>-3</sup> at  
600 10% O<sub>2</sub> for boiler < 50 kW. The tested hemp and wood fiber insulation granules do not comply  
601 with NO<sub>x</sub> emissions requirements.

602 As NO<sub>x</sub> emissions must be lowered by at least a ratio of 10 to fulfill the NO<sub>x</sub> legal threshold,  
603 a blend consisting of 10% of wood or hemp fiber and 90% of certified wood pellets was

604 suggested to be tested. The blending of hemp or wood fibers with certified wood resulted in  
 605 lower NO<sub>x</sub> emissions. Optimizing combustion conditions would reduce considerably NO<sub>x</sub>  
 606 emissions. This could be achieved via primary mechanisms (lower excess air), secondary  
 607 mechanisms (selective catalytic or non-catalytic reduction), fuel mixing (95% WR with 5%  
 608 hemp fiber or wood fiber pellets), or applying staged combustion.



609  
 610 *Fig. 6. NO<sub>x</sub> emissions as a function of the nitrogen content of the several tested biomass fuels*

611 3.2.4. Sulfur oxides emissions

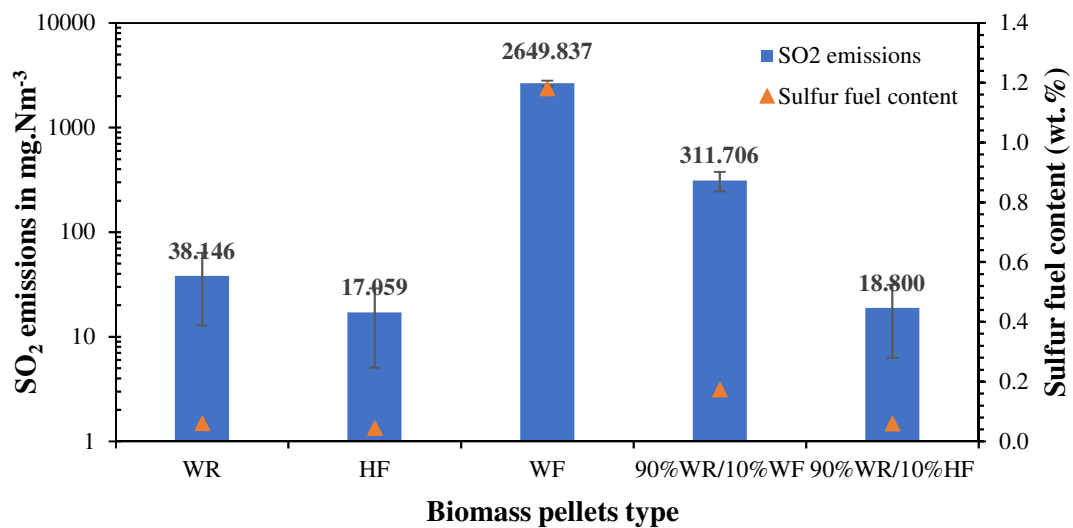
612 The sulfur content in the pellets is responsible for the formation of alkaline and alkaline-  
 613 earth sulfates (1-3%) and the generation of SO<sub>x</sub> including SO<sub>2</sub> in the majority (95%) and sulfur  
 614 trioxide in the minority (1-5%), in case of volatilization. When the flue gas is cooled in the  
 615 furnace, SO<sub>x</sub> can be transformed into sulfates and may condense on the heat exchange surfaces  
 616 leading to corrosion, or be converted into fine ash particles that cause deposit formation within  
 617 the boiler. However, SO<sub>x</sub> lead to atmospheric pollution when discharged from the stack.  
 618 Besides, SO<sub>x</sub> are proven to be toxic to humans and terrestrial animals and are implicated in the  
 619 formation of acid precipitations [59].

620 SO<sub>2</sub> emissions are directly and strongly dependent on the sulfur content of burnt fuels (Fig.  
 621 7). Therefore, the highest emissions are associated with WF (2650 mg.Nm<sup>-3</sup> having a S content



622 of 1.18 wt.% db), and the lowest emissions are associated with HF (17 mg.Nm<sup>-3</sup> having a S  
 623 content of 0.044 wt.% db). Certified wood has also quite low SO<sub>2</sub> emissions owing to its low S  
 624 content <0.06 wt.% db. When the S content of the fuel is high (> 0.2 wt.% db [30,63]), SO<sub>2</sub>  
 625 emissions become significant which could lead to boiler metal deterioration. Corrosion issues  
 626 should be anticipated throughout the WF burning.

627 As the SO<sub>2</sub> emissions from WF are extremely high, experiments involving the mixing of  
 628 WF and certified wood pellets were conducted. The SO<sub>2</sub> permissible limit of the EN 303-5:2021  
 629 standard is inexistent for small-scale boilers (< 500 kW). According to Fig. 7, the emissions of  
 630 this blend are reduced to 312 mg.Nm<sup>-3</sup> owing to the low emissions of reference wood.

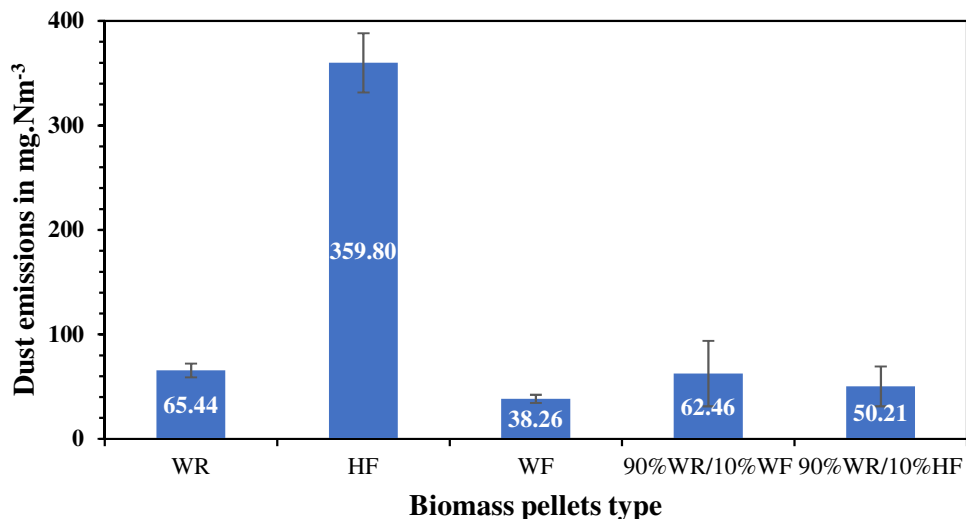


631  
 632 *Fig. 7. SO<sub>2</sub> emissions in mg.Nm<sup>-3</sup> of the different fuels and the sulfur fuel content.*

### 633 3.3. Dust emissions

634 Fig. 8 shows the total mass concentration of the particulate matter emitted during the  
 635 combustion of the different fuels and collected on the filters. These particles include mainly  
 636 unburned particles ejected from the brazier. The results are consistent with previous works  
 637 [16,21,39] in which they witnessed that the quality of the granules influences significantly  
 638 dust/PM emissions. Total PM emissions from different boilers/stoves were noticed to fluctuate  
 639 between 5 to 800 mg.Nm<sup>-3</sup> (normalized at 10% O<sub>2</sub>) [16,21,39,61,64]. In this work, the emissions

640 varied between 38 and 360 mg.Nm<sup>-3</sup> (normalized at 10% O<sub>2</sub>). These emissions were the lowest  
 641 for WF (38.26 mg.Nm<sup>-3</sup>) and the greatest for HF (359.80 mg.Nm<sup>-3</sup>). This considerably high  
 642 value for HF could be due to the formation of a significant quantity of ash and slag that could  
 643 have contributed to greater dust and fly ash emissions [65]. The mass concentration of dust  
 644 increases with the amount of potassium, chlorine, and sulfur. Reduction of up to 85% of dust  
 645 could be achieved by blending 10% HF with 90% of certified wood pellets. Compared to the  
 646 dust permissible limit indicated in EN 303-5 standard for boilers < 50 kW of class 3 (150  
 647 mg.Nm<sup>-3</sup>), class 4 (60 mg.Nm<sup>-3</sup>) and class 5 (40 mg.Nm<sup>-3</sup>), all tested pellets, except HF, satisfied  
 648 the emission requirements for class 3 and 4



649

650

*Fig. 8. Dust emissions as a function of the pellets type*

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The number concentration results of the ELPI showed that PM<sub>1</sub> (aerodynamic diameter < 1 μm), submicron particles, are dominant while larger particles with sizes from 1 to 10 μm are marginal (Table 4). Similar findings were revealed in the study conducted by Fernandes and Costa [66]. This pattern is characteristic of wood combustion. The fuel quality (ash composition), more precisely the quantity of inorganic volatiles, impacts significantly PM<sub>1</sub> emissions. PM emissions were maximum for WR (228.311×10<sup>4</sup> particles.cm<sup>-3</sup>) and minimum for HF (3.801×10<sup>4</sup> particles.cm<sup>-3</sup>). Compared to WR, the burning of HF, WF, as well as 90%WR/10%WF and 90%WR/10%HF blends reduced PM emissions by a factor of 60, 40, 42

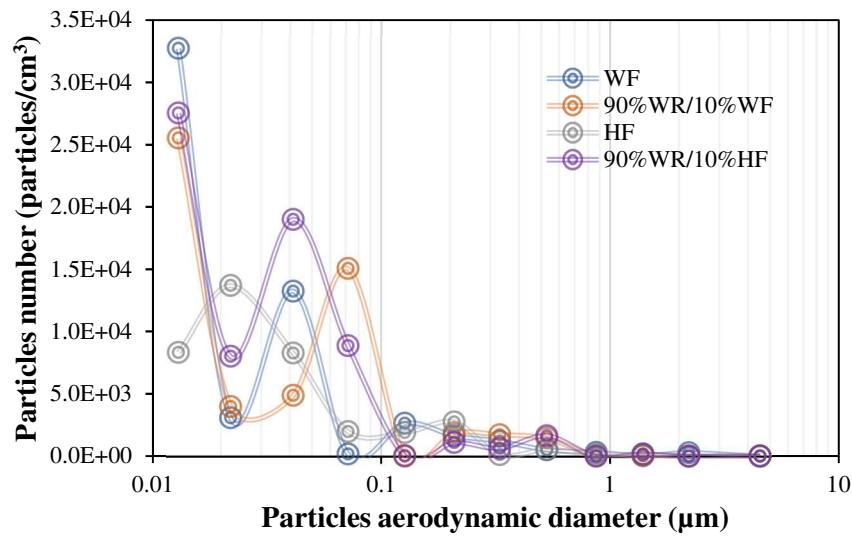
659 and 34, respectively. Pellets deriving from biomass-based insulation materials lead to  
 660 considerably lower PM number concentration, notably lower PM<sub>1</sub> emissions. These submicron  
 661 particles are considered the most toxic and difficult to separate airborne particles. Once inhaled,  
 662 the particles are deposited in the respiratory system leading to pulmonary fibrosis, chronic  
 663 obstructive pulmonary disease and asthma. Besides, these particles can induce, mutagenic,  
 664 cytotoxic, genotoxic and inflammatory responses along with oxidative stress in the human lung  
 665 cell line [67].

666 *Table 4. Particle number size distribution of the various tested pellets using ELPI*

| <b>Pellets</b>     | <b>PM<sub>1</sub> (×10<sup>4</sup><br/>particles.cm<sup>-3</sup>)</b> | <b>PM<sub>2.5</sub> (×10<sup>4</sup><br/>particles.cm<sup>-3</sup>)</b> | <b>PM<sub>10</sub> (×10<sup>4</sup><br/>particles.cm<sup>-3</sup>)</b> |
|--------------------|---|---|--|
| <b>WR</b>          | 227.559   | 228.264   | 228.311  |
| <b>HF</b>          | 3.779   | 3.800   | 3.801  |
| <b>WF</b>          | 5.558   | 5.591   | 5.591  |
| <b>90%WR/10%WF</b> | 5.452   | 5.458   | 5.461  |
| <b>90%WR/10%HF</b> | 6.684   | 6.702   | 6.704  |

667 Wood and hemp fibers along with the blending presented a bimodal PM number size  
 668 distribution with fine modes (aerodynamic diameter < 2.5 μm) (Fig. 9). WF pellets, as well as  
 669 90%WR/10%WF and 90%WR/10%HF blends, showed two ultrafine modes. The first one falls  
 670 in the nuclei modes (< 10 nm) with a mode diameter below 10 nm, while the second one falls  
 671 in the Aitken mode (10-100 nm) with mode diameters of 0.0415 μm (for WF and  
 672 90%WR/10%HF) and 0.072 μm (for 90%WR/10%WF). In these modes, the collected particles  
 673 result from gaseous low volatiles precursors (organic carbon, metal vapors, sulfuric acid, fresh  
 674 high-temperature emissions, etc.) entrained with the exhaust flue gas. The nuclei mode particles  
 675 (known also as nanoparticles) are mainly formed through homogeneous nucleation (gases  
 676 converted to vapors/particles) [68] while the Aitken mode particles (soot) are formed from the  
 677 growth of nucleation mode particles or directly emitted from biomass combustion [69]. A  
 678 slightly negligible proportion of the PM number was detected in the accumulation (100-1000  
 679 nm) and coarse (> 1000 nm) modes. The only exception was hemp fiber which showed one

680 ultrafine and one fine mode. The ultrafine mode was located between 0.0131 and 0.0415  $\mu\text{m}$   
 681 with a mode diameter of 0.0221  $\mu\text{m}$ , while the fine mode was located between 0.1274 and  
 682 0.3305  $\mu\text{m}$  with a mode diameter of 0.2095  $\mu\text{m}$ . These latter medium-size particles called also  
 683 accumulation mode particles, contain either particles that have grown through condensation and  
 684 coagulation of volatile species or directly emitted particles from combustion by the fusion of  
 685 non-volatile ash species [68,69].



686

687 *Fig. 9. Number size distribution of emitted particles during combustion of pellets derived from hemp fibers, wood fiber,*  
 688 *90%WR/10%WF and 90%WR/10%HF blends in 20 kW multi-fuel boiler under standard laboratory conditions*

689 Certified wood showed a unimodal PM number size distribution (Fig. 10). The ultrafine  
 690 mode was located between 0.0131 and 0.0415  $\mu\text{m}$  with a mode aerodynamic diameter (not  
 691 corrected for various fuel densities) of 0.0221  $\mu\text{m}$ . Therefore, fine sub-micrometer  
 692 particles/agglomerates dominate the PM resulting from the burning of certified wood, in  
 693 conformity with previous works [70,71]. Minor fractions of accumulation and coarse mode  
 694 particles were found.

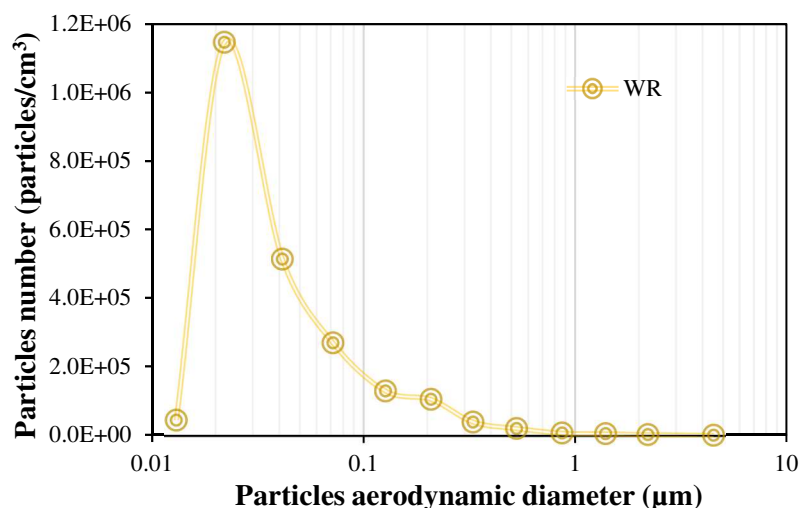


Fig. 10. Number size distribution of certified wood emitted particles during combustion in 20 kW multi-fuel boiler under standard laboratory conditions

695  
 696  
 697  
 698 The particles (ultrafine, fine and coarse) collected on the filters were having a black  
 699 color, dominated by organic matter, and soot along with a small fraction of fly ash; except for  
 700 hemp fibers which showed dust having a grey color governed by the fragmentation of HF ash.  
 701 The latter were entrained from the fuel bed on the grate and carried out with the flue-gas flow  
 702 prior to being collected on the filters [65]. The HF dust particles were mainly composed of K,  
 703 S, and Cl-containing compounds (Table 5). These inorganic species might be originated from  
 704 sulfates and alkali vapors condensation at the PM surfaces or from the incomplete vaporization  
 705 of the ash fractions [72]. These highly volatile species are present also in all the remaining  
 706 materials but at a lower concentration, as they are initially formed during the homogeneous  
 707 nucleation stage. These observations are consistent with those of Wiinikka et al. [73] and  
 708 Fernandes and Costa [74]. According to Wiinikka et al. [73], fine mode particles contained K,  
 709 S, Na, Cl and Zn while coarse mode particles consisted of refractory metals such as Ca, Mg, Si,  
 710 Mn and Fe. Similarly, Fernandes and Costa [74] have reported that the inorganic elemental  
 711 composition of the ultrafine and fine particles was dominated by mainly K, Cl, S, Ca, Mg, and  
 712 P independently of boiler operating conditions [74].

Table 5. Composition of the particle matter collected on the filters using X-ray Fluorescence

| Elements in PM (%) | WR     | HF     | WF     | 90% WR/10% WF | 90% WR/10% HF |
|--------------------|--------|--------|--------|---------------|---------------|
| <b>K</b>           | 39.618 | 76.056 | 41.555 | 32.551        | 37.157        |
| <b>Si</b>          | 20.623 | 0.223  | 14.800 | 23.039        | 20.716        |
| <b>S</b>           | 16.309 | 5.410  | 9.425  | 12.312        | 6.686         |
| <b>Ca</b>          | 7.555  | <ld    | 5.672  | 9.244         | 6.553         |
| <b>Zn</b>          | 4.882  | 2.100  | 5.856  | 7.340         | 5.075         |
| <b>Ti</b>          | 4.565  | <ld    | 5.205  | 6.732         | 4.743         |
| <b>Cl</b>          | 3.683  | 15.903 | 15.345 | 6.094         | 16.520        |
| <b>Al</b>          | 1.786  | <ld    | 1.068  | 1.433         | 1.816         |
| <b>P</b>           | 0.605  | 0.153  | 0.465  | 0.529         | 0.320         |
| <b>Fe</b>          | 0.186  | 0.077  | 0.234  | 0.261         | 0.103         |
| <b>Cr</b>          | 0.126  | 0.059  | 0.335  | 0.415         | 0.279         |

714 <ld: below the detection limit

### 715 3.4. Boiler efficiency

#### 716 3.4.1. Direct boiler efficiency

717 A comparison of the boiler efficiency operated with various tested pellets is illustrated in

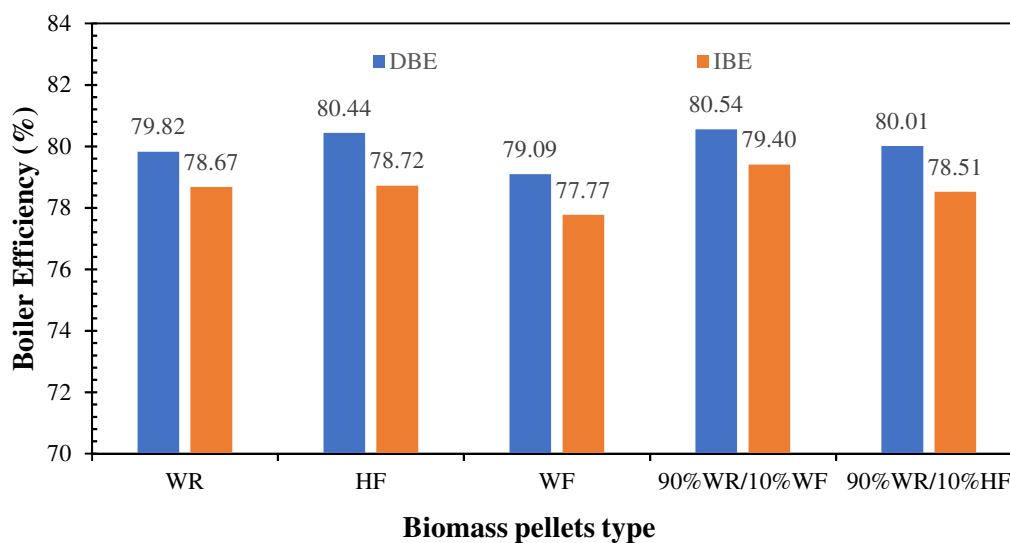
718 Fig. 11. Independently of the type of fuel burnt, the combustion efficiency of the boiler was

719 near 80% (based on HHV) and 85% (based on LHV). According to the EN 303-5 standard, the

720 minimum boiler efficiency on an LHV basis should be 74, 82 and 88% for boiler classes 3, 4

721 and 5, respectively. Therefore, the boiler used in this study satisfied the requirements for boiler

722 classes 3 and 4.



723

724  
725

Fig. 11. Direct and indirect boiler efficiency (on an HHV basis) with the various burnt pellets under standard laboratory conditions

726 3.4.2. Indirect boiler efficiency and losses

727 To better understand where the heat losses are occurring, the indirect efficiency was  
728 calculated (Fig. 11). It should be noted that this efficiency is less realistic than the direct one  
729 and reflects the efficiency of the boiler at a particular time not the overall combustion efficiency  
730 of the boiler. The indirect boiler efficiency was having the same trend as the direct efficiency,  
731 fluctuating near a slightly average lower value (78%).

732 Fig. 12 displays the magnitude of the different heat losses taking place in the boiler. The  
733 heat losses due to sensible heat of the dry flue gas ( $HL_1$ ) and due to the enthalpy of water vapor  
734 in the flue gas ( $HL_2$ ) are the key boiler losses, accounting for more than 85% of the total heat  
735 losses. The remaining losses are less significant.  $HL_1$  is the largest heat loss representing solely  
736 about 50% of the total losses. This loss is critical as it depends on the temperature of the flue  
737 gas and the combustion air flow rate. This factor relies on the excess air ratio entering the boiler  
738 that will be heated to the flame temperature. The excess air should be the lowest to avoid heating  
739 a huge amount of nitrogen that will carry out heat losses to the environment. Furthermore,  
740 reducing the flue gas temperature by using a low-temperature boiler (90-140 °C) or a  
741 condensing boiler (40-90 °C) would maximize the boiler's efficiency. For instance, if a  
742 condensing boiler is used in the same conditions stated in this work, the boiler efficiency will  
743 reach 93-97% depending on the exhaust temperature which could be set between 40 and 90 °C.

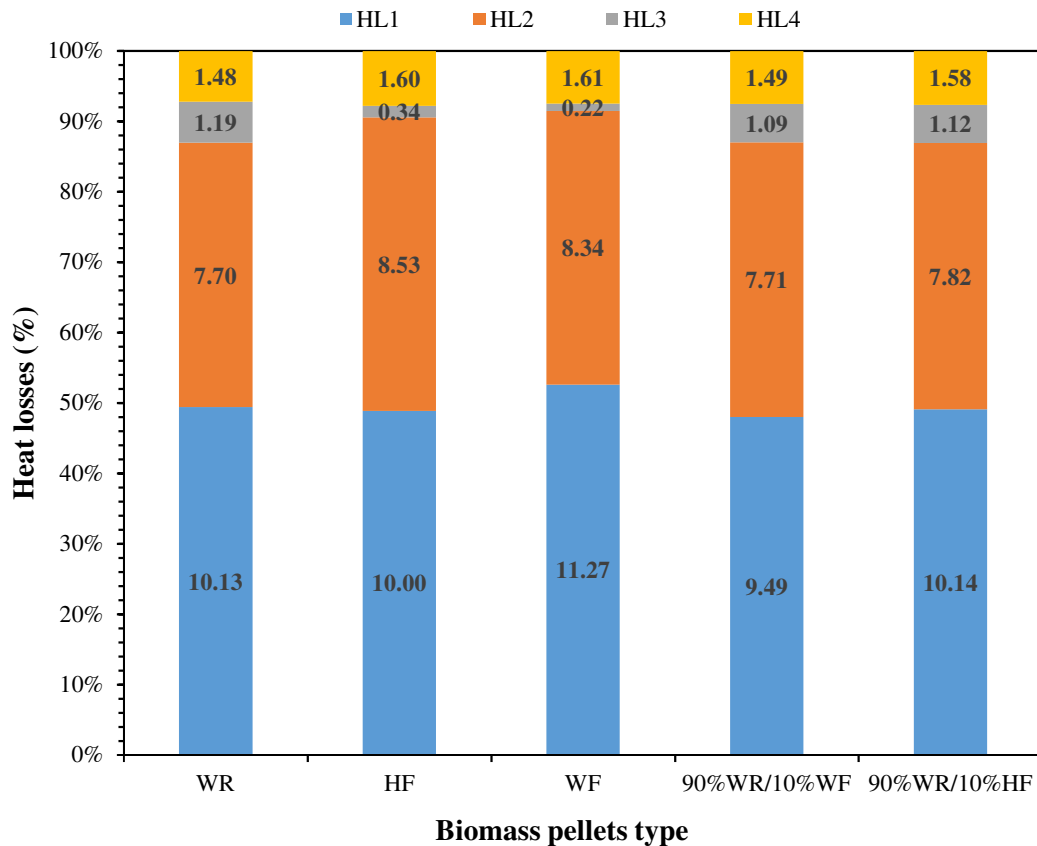


Fig. 12. Magnitude of the different calculated heat losses taking place in the boiler

### 3.5. Bottom ash composition

The melting point of the bottom ash resulting from the combustion of certified wood and blends is low. These ashes, partially or entirely melted, are deposited on the walls of the combustion chamber and hence become directly subjected to the radiative heat emanating from the flames. This deposit leads to ignition problems inside the boiler and poor combustion conditions, thus affecting the combustion performance of the boiler.

Table 6 reveals the composition of the bottom ash fraction derived from the burning of the various pellets used in this study. The ash is composed mainly of major, minor and trace inorganic elements. In declining order of abundance, major components consist of Ca, K, Si and Mg, while minor compounds comprise P, Fe, Al, Na, Mn, S, Cl and F, and trace compounds include Cu, Cr, Zn, Ni, V, As, etc. Inorganic elements vary depending on the type of biomass, its origin, the manufacturing process, and the incorporated mineral additives. Besides, these



758 elements rely on boiler operating conditions as they can be oxidized in the ash or volatilized  
759 [18]. Despite their presence in small amounts, inorganic elements can affect the combustion  
760 process.

761 The DIN $plus$  certified wood pellets presented considerably greater concentrations of iron  
762 and silica than wood and hemp fiber pellets. Typically, these values fall close to those of  
763 contaminated biomass. Soil components such as sand or clays could be the reason for the high  
764 concentration of silica in this fuel. They could have been inserted while harvesting and  
765 transporting or throughout the manufacturing of the pellets. The ash composition is consistent  
766 with the values reported in the literature for woody and non-woody fuels [61]. These ashes (rich  
767 in Ca, P, K, Fe, ...) could be used as a fertilizer for crop growth. Another application is their  
768 use in ceramic materials for the production of bricks. This could help in minimizing the  
769 extraction of raw clay and hence resulting in economic and environmental benefits due to the  
770 retention of inorganics while avoiding their landfill disposal and associated leaching concerns  
771 [75,76].

772 High chlorine content was found in HF ash. This may be attributed to the fungicide  
773 treatment based on chlorine-contained biocide. Ash resulting from wood fiber pellets and its  
774 blend (90%WR/10%WF) included the highest carbon content (11.8 and 9.5 wt.%, respectively)  
775 among the tested materials. This is due to the incorporated flame retardant (ammonium sulfate  
776  $(\text{NH}_4)_2\text{SO}_4$ ) in WF that may have delayed the combustion of the pellets and hence led to non-  
777 reacting carbon presence in the ash. This flame retardant is also behind the high sulfur and  
778 nitrogen elements found in WF ash. Studies have demonstrated that during the combustion of  
779 wood and wheat straw pellets, 40-70% and 40-55% of the sulfur is retained in the bottom ash  
780 [63,77]. The combustion temperature and the concentration of alkali (K) and alkaline earth  
781 metals (Ca and Mg) in the feedstock affect the sulfur fixation efficiency in the bottom ash  
782 fraction. Aside from being converted to  $\text{SO}_x$  in an oxidizing atmosphere, inorganic sulfur,

783 originating from ammonium sulfate, may be converted to alkaline earth metal sulfates ( $MgSO_4$   
784 and  $CaSO_4$ ) and alkali metal sulfate ( $K_2SO_4$ ) that accumulated in the solid phase [78,79]. These  
785 less corrosive sulfates have high melting temperatures, hence increasing the fusibility of the  
786 corresponding bottom ash. The melting point of the bottom ash resulting from the combustion  
787 of certified wood and blends is low. These ashes, partially or entirely melted, are deposited on  
788 the walls of the combustion chamber and hence become directly subjected to the radiative heat  
789 emanating from the flames. This deposit leads to ignition problems inside the boiler and poor  
790 combustion conditions, thus affecting the combustion performance of the boiler.

791

*Table 6. Composition of the bottom ash/slag resulting from the combustion of the different pellets*

| <b>Analytical parameters</b>                               | <b>Units</b>           | <b>WR</b> | <b>HF</b> | <b>WF</b> | <b>90% WR /10% HF</b> | <b>90% WR/ 10% WF</b> |
|--|------------------------|-----------|-----------|-----------|-----------------------|-----------------------|
| <b>Analysis of the ash fraction</b>                        |                        |           |           |           |                       |                       |
| <b>Basic analysis</b>                                      |                        |           |           |           |                       |                       |
| Ash at 815°C   | wt.% db                | 92.1      | 90.9      | 89.9      | 92.2                  | 89.0                  |
| <b>Elemental analysis</b>                                  |                        |           |           |           |                       |                       |
| Mineralization for determination of halogens and/or sulfur |                        |           |           |           |                       |                       |
| Total carbon   | wt.% db                | 5.70      | 4.10      | 11.80     | 4.90                  | 9.5                   |
| Total hydrogen   | wt.% db                | < 0.10    | 0.15      | 0.12      | < 0.10                | 0.12                  |
| Total nitrogen   | wt.% db                | < 0.10    | < 0.10    | 0.22      | < 0.10                | < 0.1                 |
| Total oxygen   | wt.% db                | 1.8       | 4.20      | < 0.20    | 2.3                   | < 0.2                 |
| Total sulfur   | mg.kg <sup>-1</sup> db | 2071      | <20       | 85057     | 3702                  | 15831                 |
| Total chlorine   | mg.kg <sup>-1</sup> db | 130       | 3484      | 574       | 414                   | 240                   |
| Total fluorine   | mg.kg <sup>-1</sup> db | < 20      | 2291      | 84        | < 20                  | < 20                  |
| <b>Metals</b>  |                        |           |           |           |                       |                       |
| Mineralization for the determination of minor metals       |                        |           |           |           |                       |                       |
| Total cadmium  | mg.kg <sup>-1</sup> Cd | < 1       | < 1       | < 1       | < 1                   | < 1                   |
| Total chromium   | mg.kg <sup>-1</sup> Cr | 258       | 16        | 142       | 182                   | 240                   |
| Total copper   | mg.kg <sup>-1</sup> Cu | 441       | 104       | 525       | 220                   | 306                   |
| Total aluminum   |                        | 19951     | 4710      | 17252     | 19633                 | 13600                 |
| Total iron   |                        | 19510     | 6154      | 10280     | 14755                 | 15105                 |
| Total manganese  |                        | 6446      | 2402      | 5878      | 3792                  | 4424                  |
| Total magnesium  | mg.kg <sup>-1</sup> db | 30999     | 46197     | 45655     | 39684                 | 35100                 |
| Total potassium  |                        | 82937     | 209044    | 65088     | 115564                | 82937                 |
| Total phosphorus   |                        | 15241     | 31006     | 13363     | 14629                 | 9738                  |
| Total sodium   |                        | 10461     | 2448      | 4600      | 7864                  | 10461                 |
| Total calcium  |                        | 221557    | 230205    | 208335    | 182177                | 161808                |
| Total nickel   | mg.kg <sup>-1</sup> Ni | 60        | < 1       | 59        | 48                    | 49                    |
| Total silicium   | mg.kg <sup>-1</sup> db | 131555    | 16409     | 37353     | 141372                | 153995                |
| Total lead   | mg.kg <sup>-1</sup> Pb | 3         | < 1       | 2         | 9                     | 7                     |
| Total vanadium   | mg.kg <sup>-1</sup> V  | 31        | 10        | 15        | 30                    | 27                    |
| Total zinc   | mg.kg <sup>-1</sup> Zn | 62        | 47        | 117       | 109                   | 118                   |

|  |                        |       |        |       |       |       |
|--|------------------------|-------|--------|-------|-------|-------|
| Total arsenic  | mg.kg <sup>-1</sup> As | 9     | < 1    | 5     | 13    | 15    |
| Total mercury  | mg.kg <sup>-1</sup> db | < 0.1 | < 0.1  | < 0.1 | < 0.1 | < 0.1 |
| <b>Ash fusibility</b>  |                        |       |        |       |       |       |
| <b>Fusibility of ash in an oxidizing atmosphere at 815°C</b> |                        |       |        |       |       |       |
| Initial shrinkage temperature                                | °C                     | 1100  | 1410   | 1005  | 1100  | 1140  |
| Deformation temperature                                      | °C                     | 1243  | > 1500 | 1337  | 1204  | 1202  |
| Hemisphere temperature                                       | °C                     | 1266  | > 1500 | 1437  | 1216  | 1216  |
| Flow temperature   | °C                     | 1282  | > 1500 | 1477  | 1230  | 1238  |

#### 4. Conclusions

The combustion and emission characteristics of certified wood pellets, hemp-fiber, and wood-fiber insulation panels waste pellets along with the blends (certified wood/hemp fiber, certified wood/wood fiber) have been examined when fired in a small-scale multi-fuel boiler (20 kW) under standard laboratory conditions. Initially, the insulation panel wastes were shredded and pelletized. To assess the potential of using waste pellets from a sustainable origin in domestic boilers, the resulting pellets were then characterized and the values were compared to their corresponding standards (certified straw and wood pellets). The following major conclusions could be withdrawn from this work:

- Hemp-fiber pellets satisfied all the European standard requirements for non-industrial pellets.
- Wood fiber pellets could not meet all the permissible values related to their elemental composition (high nitrogen and sulfur contents).
- The use of pellets originating from biomass-based building insulation materials impacted considerably the gaseous and particulate matter emissions. Among the fuels, wood fiber pellets have emitted the highest concentration of nitrogen and sulfur oxides owing to the high proportion of nitrogen and sulfur in the material. Furthermore, The mass concentration of particulates was the lowest for these pellets (38 mg.Nm<sup>-3</sup>). However, the burning of hemp fiber pellets resulted in high nitrogen oxide emissions, the lowest sulfur oxide emanations and the highest mass concentration of particle

812 matter ( $360 \text{ mg.Nm}^{-3}$ ). These particulate emissions were dependent on the ash content  
813 of the raw material and composition; namely the volatile inorganic fly ash elements K,  
814 Si, S and Cl. For all tested feedstock, submicron particles ( $< 1 \mu\text{m}$ ) were dominant ( $10^4$   
815  $-10^6 \text{ particles.cm}^{-3}$ ); with significantly lower fine particles emitted by the burning of  
816 hemp and wood fiber pellets compared to certified wood pellets.

817 • The blends 90%WR/10%WF and 90%WR/10%HF satisfied all the emissions  
818 requirements of the EN ISO 303-5 standard (except CO and  $\text{NO}_x$  emissions).  
819 Combustion conditions have to be tuned and optimized to mitigate CO and  $\text{NO}_x$   
820 formation. CO emissions could be reduced by providing higher excess air in the  
821 combustion chamber for better mixing of the fuel with air, operating at higher furnace  
822 temperatures and increasing the residence time. On the other hand,  $\text{NO}_x$  reduction could  
823 be achieved via primary mechanisms (lower excess air), secondary mechanisms  
824 (selective catalytic or non-catalytic reduction) or fuel mixing (95% WR with 5% hemp  
825 fiber or wood fiber pellets).

826 • The use of pellets originating from biomass-based building insulation wastes affected  
827 marginally the boiler efficiency. The flue gas losses were governing the heat losses  
828 taking place in the boiler for all tested fuels. The optimization of the excess air level  
829 could minimize the CO emissions and stack losses. Using a low-temperature  
830 condensing boiler will help in maximizing the boiler efficiency ( $> 93\%$ ) while  
831 minimizing the sensible and latent heat losses.

832 • Blending biomass-based insulation materials waste with certified wood pellets has an  
833 attractive potential to be used as renewable substitute feedstock to answer the future  
834 demand for pellets and reduce the French importation of pellets for residential boilers.  
835 Using blends can also help in using the same boiler conditions tuned for certified wood  
836 while reducing substantially the gaseous and particulate matter emissions. The blends

837 will allow the expansion of the spectrum of biomass raw materials used in small-scale  
838 combustion systems and allow the introduction of new biomass waste (such as hemp  
839 fiber and wood fiber insulation panels) that potentially may end up in landfills.

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## 846 Data availability

847 The data related to this article are not publicly available at the present moment but can be  
848 provided by the authors upon reasonable request

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**Biomass-based  
Insulation  
materials**



**Pellets**



**Hemp fibers**



**Wood fibers**

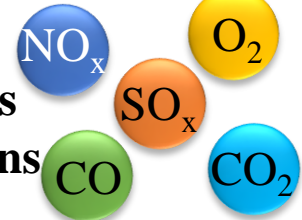


**Commercial Wood**



**Boiler efficiency (%)  
+ Heat Losses (%)**

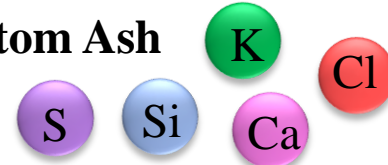
**Gaseous  
emissions**



**Dust emissions:  
Mass concentration  
+ size distribution**



**Bottom Ash**



**Boiler efficiency**

**HL1**

**HL3**

**Heat Losses**

**HL2**

