

PARTICLE NUMBER EMISSIONS AND SIZE DISTRIBUTIONS FROM RESIDENTIAL WOOD STOVES UNDER LABORATORY CONDITIONS

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Abstract

The objectives of the study were to experimentally investigate particle number emissions and number size distributions from two modern residential wood stoves with capacity 10 kW and 20 kW under laboratory conditions using a state of the art instrument Electrical Low Pressure Impactor Plus (ELPI+). The ELPI+ measures particles in real time with a fast response time in a wide particle size range from 6 nm to 10 µm aerodynamic diameter. Five combustion batches for each wood stove were conducted for the particle emission measurements in a laboratory of a stove manufacturing plant in Namur, Belgium. Batch 5 for the 10kW stove was operated in part load heat output. The combustion experiments were conducted with minor modification of primary air supply into the combustion chamber. Experimental results show that particle number concentrations measured from both stoves varied between 3.4×10^7 to 8.7×10^7 particles/cm³ for the startup phase, 2.6×10^7 to 5.0×10^7 particles/cm³ for the combustion phase, 1.8×10^7 to 2.7×10^7 particles/cm³ for the burnout phase and 2.7×10^7 to 5.3×10^7 particles/cm³ for the total cycle. The startup phase of all the batches was characterized by relatively higher particle number emissions probably due to low temperature condition and dust carryover. Particle number size distribution results show that the majority of all the phases has bimodal size distributions with one mode has a peak particle number concentrations at the particle size around 20 nm while another mode has maximum concentrations at the particle size between 70 nm to 125 nm.

Keywords : Biomass combustion, wood stoves, batch combustion, particle number emissions, number size distributions

Introduction

The usage of biomass fuel is increasing across the globe because it is a renewable energy source. Biomass is widely available worldwide and provides a major share of renewable energy in most countries. It is considered as one of the options to mitigate greenhouse gas emissions and substitute

fossil fuels (Obaidullah, 2014; Obaidullah *et al.*, 2014; Obaidullah *et al.*, 2018a; 2018b; 2018c). It is used for power and heat energy needs across Europe, heating people's homes directly or through district heating and providing power to business and homes. The Renewable Energy Directive

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(2009/28/EC) sets a mandatory target for the Member States in the EU (European Union) countries to increase their share of renewable energy sources and fuels to 20% by 2020 (Scarlat *et al.*, 2015). Sustainable wood and wood products accounted for almost 45% of the EU's gross inland final energy consumption of renewables in 2016 and the renewable energy contribution is further expected to increase to 55-75% in 2050 (Scarlat *et al.*, 2015). Biomass combustion in small scale heating appliances is an important source of carbonaceous particle emissions in the EU countries, and makes new challenges to both indoor and outdoor air quality, human health, and global and regional climate (Bolling *et al.*, 2009; Vicente and Alves, 2018). These appliances are accused of producing harmful particulate matter (PM) pollutants, since they generate significant amounts of fine particle (size less than 1 μm) emissions at low stack height and these smaller particles emitted directly to the ambient air. Residential biomass combustion is a major source of primary fine organic particles and secondary organic particles.

Biomass combustion produces large quantities of fine particulate matter that have a negative impact on ambient air quality (Obaidullah *et al.*, 2018b). This could be harmful especially in densely populated areas. Recent investigations have reported that small scale residential biomass combustion was responsible for more than 45% of total $\text{PM}_{2.5}$ (particle size smaller than 2.5 μm) emissions in Europe in 2015 (Heat, 2016; EEA, 2018). Several studies have mentioned that increased particle concentrations in the ambient air correlate with adverse health effects in the exposed population, including respiratory and cardiovascular illnesses as well as increased mortality (Lighty *et al.*, 2000; Hackley *et al.*, 2007; Bolling *et al.*, 2009; Polichetti *et al.*, 2009; Bari *et al.*, 2011; Obaidullah *et al.*, 2012a; 2012b; 2012c; 2012d; 2013; Obaidullah and De Ruyck, 2020). Smaller size particles ($\text{PM}_{2.5}$) are considered more dangerous for human health because they can travel deeper in the alveolar region of the respiratory tract. The combustion particles also cool the atmosphere by scattering sunlight and the particles form clouds that back sunlight to the space (Sippula, 2010; Obaidullah *et al.*, 2012a; Obaidullah, 2014).

Residential heating appliances that use biomass fuel are usually classified into two main categories : batch fired appliances and continuously fired appliances. Batch fired appliances are less efficient than the appliances equipped with continuous combustion. Traditional domestic biomass heating appliances emit significant amount of pollutants, especially PM emissions (Obaidullah, 2014). Small scale heating appliances such as wood stoves and

fire places are very common for space heating. There are several types of heating appliances in households equipped with a vast range of different combustion technologies. Due to differences in combustion technologies and type of applications, the combustion conditions will vary between different appliances and combustion phases and this has a major impact on emissions.

A number of studies on particle emission from small scale biomass combustion heating appliances were found in the literature. Bari *et al.* (Bari *et al.*, 2011) investigated particle number emissions from a pellet stove (5 kW) and a wood log boiler (20 kW). The measurements were done from the stack using a Scanning Mobility Particle Sizer (SMPS) for number concentrations and number size distributions. Particle number concentrations varied between 1.2×10^7 and 4.7×10^7 particles/ cm^3 for pellet stove, while 1.5×10^7 and 5.4×10^7 particles/ cm^3 for the wood log boiler. They observed that number size distributions of emitted particles were unimodal in the size range of 60 nm to 100 nm. Sippula O. (Sippula, 2010) examined fine particle emissions from a top feed pellet stove with capacity of 8 kW. An Electrical Low Pressure Impactor (ELPI) was used to measure particle emissions characteristics after the dilution tunnel. From his results, it was observed that particle number concentrations varied between 1.1×10^7 and 4.7×10^7 particles/ cm^3 . Johansson *et al.* (Johansson *et al.*, 2003) investigated particle number emissions from a domestic pellet stove with a capacity of 6 kW. The stove was fired with wood pellets. Particle characterizations were done with an ELPI. Experimental results show that particle number concentrations varied between 1.8×10^7 and 8.7×10^7 particles/ cm^3 . Gaegauf *et al.* (Gaegauf *et al.*, 2001) studied particle number emission and number size distributions from a wood pellet boiler (17 kW) and a wood log boiler (25 kW). An SMPS was used to determine particle number concentrations and size distributions from the stack. They observed that particle number concentrations were 0.8×10^7 particles/ cm^3 for the pellet boiler and 1.8×10^8 particles/ cm^3 for the wood log boiler. The number size distribution results show that the major part of the particle number emissions is in the range between 30 and 300 nm. Ozgen *et al.* (2013) investigated particle number emissions and size distributions from biomass fired in a closed fireplace. An EPLI was used to characterize particle concentrations with a two stage dilution tunnel. The results show that average particle number concentrations from the study were 8.7×10^8 particles/ cm^3 . Altaher *et al.* (2015) showed that the particle number emissions from biomass pellet combustion had a particle size distributions in the nano-particle range with

a peak number concentrations 30 nm of 5×10^8 particles/cm³.

The above review illustrates particle number emissions mainly for the combustion phase of combustion cycle. Generally, the combustion cycle in batch fired wood stoves is characterized by the startup, the combustion and the burnout phases. It is well known that amounts of particle number emissions from different phases of batch fired stoves differ generally. However, the enough information on characterizing the particle number emissions from different phases of the combustion cycle is usually not found in the literature. Besides, it can be seen from the above mentioned review that limited research on particle emission from batch fired combustion especially wood stoves have been investigated. There is a need for much knowledge on particle number emissions and size distributions from wood stoves in different phases. The objectives of the present work were to experimentally evaluate particle number emissions and number size distributions from two commercially available modern wood stoves with a capacity 10 kW and 20 kW respectively. A total of 10 combustion batches from both wood stoves were conducted for the particle number emission measurements at the stove manufacturing plant in Belgium. The work includes particulate number emissions and number size distributions from different phases of the combustion cycle as well as a comparison with previously reported data.

Materials and Methods

Fuel Properties

The chemical composition of the wood log fuels was tested in the laboratory of SGS Environmental Services, Arnhem, the Netherlands. The wood log was Beech species available in the forest of Belgium. The properties of wood log fuel used in the combustion experiments of the 10 kW and 20 kW wood stoves are presented in Table 1.

Combustion Appliances

The combustion appliances used in the experiments were two different commercially available Belgian made modern wood stoves of 10 kW and 20 kW heat output respectively. Both appliances are chimney type free standing logwood stove for manual operation. For reaching the nominal output of the 10 kW stove, 2.2 kg of logwood (2 pieces) is combusted in every batch. The volume of the combustion chamber is approximately 0.090 m³. For reaching the nominal output of the 20 kW stove, about 4.4 kg of logwood (4 pieces) is combusted per batch. The volume of the combustion chamber is 0.175 m³. The appliance is equipped with four convection outputs. The initial firing in the wood stoves was performed with a small ignition fire consisting of thin wood sticks together with paper. Preheating of the stoves was performed before starting the measurements of the combustion batches. After ignition, the combustion device was heated up in two subsequent batches by burning 5 kg of wood log (2.5 kg per batch) for the 10 kW stove and 10 kg of wood log (5 kg per batch) for the 20 kW stove. The flue gas temperature gradually reached a stable temperature of about 280°C within an hour. Wood log was put horizontally on the combustion chamber of both stoves. The flame is directed upwards and can be seen from the outside of the stoves, since the front side of the stoves has a transparent glass window. The top surface of the combustion chamber is equipped with a baffle plate made of vermiculite with high temperature insulation material. The other sides of the combustion chamber are insulated with refractory ceramic bricks made of calcium silicate. Primary combustion air is supplied through the bottom of the combustion chamber and secondary air through the holes in the back wall approximately at middle of the height. The primary air can be adjusted by the control valve placed on the front of the appliance. The heat generated in the stoves during combustion is delivered to the surrounding space by convection and radiation from the hot

Table 1. Properties of the wood log fuel used in the combustion experiments

Parameter	Unit	Results	Analysis
Length	cm	33	Physical
Diameter	cm	10	
Weight	kg	1.15	
LHV	MJ/kg	16.2	Proximate
Moisture	% of fuel weight	13	
Ash	% of dry weight	0.3	
C (wt %)	% of fuel weight	43.8	Ultimate
H (wt %)	% of fuel weight	4.8	
O (wt %)	% of fuel weight	38.3	

surfaces of the stoves and stove pipes. In the combustion experiments, the stove was situated on a balance to allow for the measurement of the fuel mass consumption. The combustion gases were led through an externally insulated steel stack of 180 mm in diameter. Both stoves work with natural draft and can also be controlled by using an exhaust fan placed at the top of the chimney. Temperature of the indoor air and flue gas in the stack was measured by K type thermocouples.

Experimental Setup

The schematic of the experimental setup for the particle number emission measurements is shown in Figure 1. The measurement protocols of each combustion test were conducted according to the European standard EN 13229 (EN-13229, 2002). For each stove, combustion experiments were repeated for five batches to get a clear picture of the particle number emissions results. Table 2 presents fuel load, valve opening position of primary air flow into the combustion chamber and dilution air flow into the dilution tunnel carried out in experimental batches of the 10 kW stove and 20 kW wood stove.

Particle Sampling and Dilution Ratio

For particle number measurements, about above the wood stoves, a partial flow from the stack was led through an externally insulated bent tube probe which was made of stainless steel 12 mm diameter. The opening of the probe was pointed towards against the flow of the flue gases. The flue gases were passed through a partial flow dilution tunnel before reaching the particle measuring instrument, the Electrical Low Pressure Impactor Plus (ELPI+). The dilution tunnel consists of a porous tube diluter (PRD), an ejector diluter (ED) and an air heater. The first dilution takes place in the PRD, where 17.5 LPM of preheated 150°C dilution air passes through the pores of the tube while the

raw sample flows through the tube. Then 49 LPM of secondary dilution air is injected into the ejector diluter. Two mass flow controllers (Aalborg brand, Germany) are placed before the PRD and ED to regulate the flow of dilution air. The dilution air was filtered through a Festo air filtration system (a Belgian company) with four stages: Stage-1 is a condensate separator where water is removed from the dilution air stream. Stage-2 is a pre-filter where

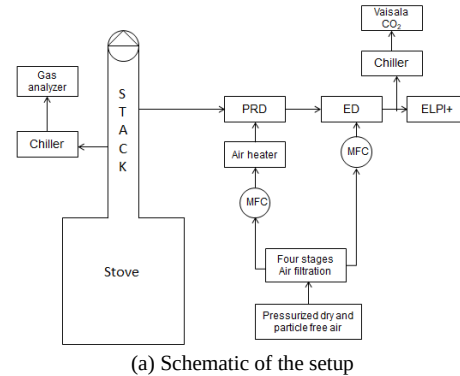


Figure 1. Experimental setup for emission measurements from wood stoves, conducted in a laboratory, Namur, Belgium, PRD = Porous tube diluter, ED = Ejector diluter, MFC = Mass flow controller

Table 2. Fuel load, valve opening position and dilution air carried out in different batches of the 10 kW stove and 20 kW stove

No. of Batches	Fuel load (kg)	Valve opening position of primary air flow (%)	Dilution air flow into PRD (LPM)	Dilution air flow into ED (LPM)
10 kW stove				
Batch 1	2.21	20	17.5	49
Batch 2	2.21	30	17.5	49
Batch 3	2.21	25	17.5	49
Batch 4	2.14	25	17.5	49
Batch 5	0.90	25	17.5	49
20 kW stove				
Batch 1	4.40	10	17.5	49
Batch 2	4.40	12	17.5	49
Batch 3	4.35	15	17.5	49
Batch 4	4.40	15	17.5	49
Batch 5	4.37	15	17.5	49

PRD = Porous Tube Diluter, ED = Ejector Diluter, LPM = Liters Per Minute

coarse particles are removed. Stage-3 is a chemical filter where hydrocarbons and oils are removed. Finally, Stage-4 is a post-filter that removes fine particles from the dilution air stream. After the filtration, average total particle number concentration of the dilution air was of 4.7×10^2 particle/cm³, entered the two stage dilution tunnel. The outlet of the ejector diluter, with the diluted sample, was connected to the particle measurement instrument ELPI+ by a flexible conductive silicon tube about 80 cm long. This material reduces the build-up of static charge and minimizes particle losses to the walls of the tube.

Particle number concentrations and size distributions were measured in real time using an Electrical Low Pressure Impactor Plus (ELPI+) from Dekati Ltd., Finland. It measures particles with a fast response time in a wide particle size range from 6 nm to 10 µm aerodynamic diameter. Samples particles enter the ELPI+ are first charged in the charger. After being charged, the particles are introduced in the 14 stage cascade impactors in order to be separated on the basis of their inertia. The charged particles collected in each impactor stage produce an electrical current, which is recorded by the respective electrometer channel. This current is proportional to particle number concentrations via mathematical algorithms (Marjamäki *et al.*, 2000). A vacuum pump with a flow rate of 10 LPM is connected at the downstream of the ELPI+ for the suction of the diluted sample flow into the ELPI+ for PM analysis. Aluminium foils with a diameter of 25 mm and thickness of 0.1 mm were placed on each impactor stage. Aluminium substrates greased Apiezon-L with a mixture of acetone were used.

The dilution ratio (DR) was calculated based on concentration of CO₂ in the raw sample and the

diluted gas sample using the following equation (Obaidullah, 2014; Obaidullah *et al.*, 2015; 2018a; 2019a; 2020).

$$DR = \frac{CO_{2,RG} - CO_{2,BG}}{CO_{2,S} - CO_{2,BG}}$$

Where, CO_{2, RG} is the concentration in the raw gas, CO_{2, BG} is the background dilution air and CO_{2, S} is the diluted sample. The CO₂ concentrations of the raw sample and the diluted sample were continuously monitored and recorded, in order to determine the prevailing dilution ratio, using a Horiba PG-250 and a Vaisala Carbocap GMP-343 analyzers respectively. The measurement principle of both analyzers is non-dispersive infrared (NDIR). Portable gas conditioning systems (M&C, Germany) were placed on the upstream of each analyzer to separate condensate water from the raw exhaust gases and diluted sample.

Results and Discussion

As the objective of this study was to evaluate the particle number emissions from different combustion phases of each batch, the emission results from each batch are arbitrary presented as the startup, the combustion and the burnout phases based on the stack temperature and the flame visualization criteria. The beginning of the combustion phase was characterized by observing the relative stabilization of the stack temperature and a strong visible flame. The emissions from the burnout phase were not recorded after every batch. At the end of each measurement day, the measurements for the burnout phase were

Table 3. Average CO₂, O₂, excess air (λ), dilution ratio and particle number concentrations with standard deviation (±) measured from the 10 kW wood stove

No. of Batches	Phase	Duration (min:sec)	O ₂ (vol %)	CO ₂ raw sample (vol %)	CO ₂ diluted sample (ppm)	Flue gas temp (°C)	Excess Air (λ)	η (%)	DR	Number (particles/cm ³)
Batch 1	SP	5:45	9.2±0.9	11.4±1.1	3468±25	263±13	1.79	-	36.6±1.5	$6.6 \times 10^7 \pm 9.2 \times 10^6$
	CP	39:45	9.7±0.8	10.5±0.7	3386±146	294±8	1.87	78.9	35.3±3.7	$4.3 \times 10^7 \pm 4.4 \times 10^6$
	Total	45:30	9.7±0.8	10.6±0.8	3397±182	288±15	1.86	-	35.5±3.4	$4.7 \times 10^7 \pm 5.4 \times 10^6$
Batch 2	SP	5:45	9.1±1.1	11.8±1.2	3257±348	284±8	1.76	-	41.7±1.9	$5.9 \times 10^7 \pm 1.8 \times 10^7$
	CP	36:45	8.0±0.5	12.1±0.6	3588±138	305±7	1.62	79.8	37.2±2.7	$5.0 \times 10^7 \pm 5.2 \times 10^6$
	Total	41:45	8.1±0.7	12.0±0.7	3549±195	301±11	1.64	-	37.8±2.8	$5.2 \times 10^7 \pm 9.9 \times 10^6$
Batch 3	SP	5:15	9.3±1.0	11.5±1.1	3090±208	295±7	1.80	-	42.9±1.9	$4.9 \times 10^7 \pm 7.1 \times 10^6$
	CP	36:00	8.1±0.6	12.4±0.5	3580±82	308±3	1.64	80.2	38.9±2.1	$4.5 \times 10^7 \pm 4.9 \times 10^6$
	Total	40:15	8.3±0.7	12.3±0.7	3519±124	305±7	1.66	-	40.9±2.2	$4.7 \times 10^7 \pm 5.7 \times 10^6$
Batch 4	SP	6:00	10.0±1.1	10.7±1.2	3569±287	278±7	1.92	-	32.9±3.3	$3.6 \times 10^7 \pm 4.4 \times 10^6$
	CP	42:15	9.7±0.4	10.7±0.4	3500±86	292±4	1.87	79.1	34.3±1.5	$3.2 \times 10^7 \pm 2.7 \times 10^6$
	Total	47:15	9.7±0.6	10.7±0.7	3507±136	290±8	1.87	-	33.6±1.8	$3.3 \times 10^7 \pm 3.2 \times 10^6$
Batch 5 (Part load)	SP	5:00	10.9±1.0	9.2±1.1	3136±298	244±11	2.10	-	33.1±1.5	$3.2 \times 10^7 \pm 4.2 \times 10^6$
	CP	34:45	11.2±0.4	8.9±0.4	3168±51	258±11	2.16	78.3	36.6±1.3	$2.8 \times 10^7 \pm 2.6 \times 10^6$
	Total	39:45	15.6±0.4	4.4±0.4	1927±109	255±9	2.14	-	34.9±1.4	$2.9 \times 10^7 \pm 3.2 \times 10^6$
	BP	30:00	11.1±0.6	9.0±0.6	3164±121	-	3.97	-	28.8±1.1	$1.8 \times 10^7 \pm 1.4 \times 10^6$

SP = startup phase, CP = combustion phase, BP = burnout phase, PAF = primary air flow, DR = dilution ratio, η = combustion efficiency

Table 4. Average CO₂, O₂, excess air (λ), dilution ratio and particle number concentrations with standard deviation ($\pm$) measured from the 20 kW wood stove

No. of Batches	Phase	Duration (min:sec)	O ₂ (vol %)	CO ₂ raw sample (vol %)	CO ₂ diluted sample (ppm)	Flue gas temp (°C)	Excess Air (λ)	η (%)	DR	Number (particles/cm ³)
Batch 1	SP	5:00	11.8 $\pm$ 0.9	8.7 $\pm$ 0.8	2,568 $\pm$ 214	277 $\pm$ 11	2.29	-	38.9 $\pm$ 2.6	8.7 $\times$ 10 ⁷ $\pm$ 6.4 $\times$ 10 ⁷
	CP	46:00	10.9 $\pm$ 0.4	9.2 $\pm$ 0.5	2,864 $\pm$ 99	291 $\pm$ 10	2.10	76.2	36.6 $\pm$ 1.2	4.2 $\times$ 10 ⁷ $\pm$ 4.2 $\times$ 10 ⁶
	Total	51:00	11.0 $\pm$ 0.5	9.2 $\pm$ 0.6	2,835 $\pm$ 138	289 $\pm$ 11	2.12	-	36.9 $\pm$ 1.7	4.7 $\times$ 10 ⁷ $\pm$ 2.2 $\times$ 10 ⁶
Batch 2	SP	5:00	11.8 $\pm$ 0.8	8.5 $\pm$ 0.7	2,525 $\pm$ 226	264 $\pm$ 10	2.29	-	40 $\pm$ 2.4	4.4 $\times$ 10 ⁷ $\pm$ 1.8 $\times$ 10 ⁷
	CP	43:15	10.6 $\pm$ 0.5	9.5 $\pm$ 0.5	2,907 $\pm$ 110	298 $\pm$ 6	2.04	76.4	37.3 $\pm$ 1.6	4.0 $\times$ 10 ⁷ $\pm$ 3.4 $\times$ 10 ⁶
	Total	48:15	10.7 $\pm$ 0.6	9.4 $\pm$ 0.6	2,869 $\pm$ 159	294 $\pm$ 12	2.06	-	37.6 $\pm$ 1.9	4.2 $\times$ 10 ⁷ $\pm$ 8.1 $\times$ 10 ⁶
Batch 3	SP	5:00	9.3 $\pm$ 1.1	11.2 $\pm$ 1.1	2,920 $\pm$ 269	290 $\pm$ 15	1.80	-	45 $\pm$ 2.9	4.8 $\times$ 10 ⁷ $\pm$ 7.4 $\times$ 10 ⁶
	CP	38:00	10.4 $\pm$ 0.3	9.8 $\pm$ 0.3	2,925 $\pm$ 91	319 $\pm$ 7	1.99	75.9	37.9 $\pm$ 1.0	2.7 $\times$ 10 ⁷ $\pm$ 2.8 $\times$ 10 ⁶
	Total	43:00	9.9 $\pm$ 0.6	10.3 $\pm$ 0.7	2,903 $\pm$ 156	315 $\pm$ 13	1.90	-	29.5 $\pm$ 0.8	3.7 $\times$ 10 ⁷ $\pm$ 4.3 $\times$ 10 ⁶
	BP	50:00	16 $\pm$ 0.5	3.8 $\pm$ 0.4	1,671 $\pm$ 53	-	4.23	-	38.9 $\pm$ 1.3	2.4 $\times$ 10 ⁷ $\pm$ 1.7 $\times$ 10 ⁶
Batch 4	SP	6:30	12.7 $\pm$ 1.0	7.8 $\pm$ 1.1	3,309 $\pm$ 175	252 $\pm$ 8	2.54	-	25.4 $\pm$ 2.6	3.8 $\times$ 10 ⁷ $\pm$ 1.4 $\times$ 10 ⁷
	CP	45:00	10.6 $\pm$ 0.3	9.7 $\pm$ 0.4	3,511 $\pm$ 116	273 $\pm$ 7	2.03	78.3	31.0 $\pm$ 0.9	3.7 $\times$ 10 ⁷ $\pm$ 2.3 $\times$ 10 ⁶
	Total	51:30	10.8 $\pm$ 0.5	9.5 $\pm$ 0.5	3,491 $\pm$ 133	271 $\pm$ 10	2.07	-	30.4 $\pm$ 1.4	2.9 $\times$ 10 ⁷ $\pm$ 6.4 $\times$ 10 ⁶
Batch 5	SP	5:00	11.5 $\pm$ 1.0	9.1 $\pm$ 1.1	3,364 $\pm$ 293	263 $\pm$ 7	2.22	-	29.3 $\pm$ 2.5	3.4 $\times$ 10 ⁷ $\pm$ 5.3 $\times$ 10 ⁶
	CP	42:45	11.0 $\pm$ 0.3	9.2 $\pm$ 0.3	3,325 $\pm$ 98	279 $\pm$ 4	2.12	77.3	31.0 $\pm$ 0.7	2.6 $\times$ 10 ⁷ $\pm$ 2.5 $\times$ 10 ⁶
	Total	47:45	11.1 $\pm$ 0.6	9.2 $\pm$ 0.6	3,329 $\pm$ 173	277 $\pm$ 8	2.13	-	25.8 $\pm$ 0.8	2.1 $\times$ 10 ⁷ $\pm$ 1.1 $\times$ 10 ⁶
	BP	22:00	14.9 $\pm$ 0.4	4.9 $\pm$ 0.4	2,271 $\pm$ 62	-	3.46	-	30.8 $\pm$ 1.3	2.7 $\times$ 10 ⁷ $\pm$ 3.3 $\times$ 10 ⁶

SP = startup phase, CP = combustion phase, BP = burnout phase, PAF = primary air flow, DR = dilution ratio, η = combustion efficiency

performed. Three burnout phases were conducted in the campaign of the particle emission measurements. The particle emission results for each phase measured from the partial flow dilution tunnel were corrected by multiplying the corresponding dilution ratio (DR). Since the combustion process varies significantly over time in the batch fired heating appliances, the presented standard deviation ($\pm$) also describes the variation of the process. The particle number emission results together with the DR and the combustion efficiency of the 10 kW stove and 20 kW stove are presented in Tables 3 and 4 respectively. It can be seen from Tables 3 and 4 that the combustion efficiency from all the batches was 79.3 $\pm$ 0.7% for the 10 kW stove and 77.1 $\pm$ 1.2% for the 20 kW stove.

Time Series of Oxygen Concentrations, Dilution Ratio and Number Concentrations

The combustion process in residential wood stoves is characterized by three combustion phases: the startup (ignition) phase, the combustion phase and the burnout phase. The startup phase is generally showing high concentrations of particle emissions. At the beginning of a combustion cycle during this phase, the concentration of O₂ in the flue gas decreases while the temperature in the furnace starts to increase. The startup phase of the stoves takes about 5 to 7 min to complete. As soon as stable combustion conditions have been reached, the combustion phase begins. This phase is characterized by low particle emissions in the flue gas and a higher temperature, which provides better combustion conditions. The combustion phase takes about 40 to 46 min to complete. At the end of the combustion cycle, charcoal burnout

phase takes place. O₂ concentrations in the flue gas start to increase and the furnace temperature decreases. This phase takes about 25 to 30 min to complete. The time for supplying a new batch of wood log was defined from the moment when no visible flames were observed and CO₂ concentration reached about 5 %. Then, a batch of wood log was supplied on the glow bed of the previous fire.

Figure 2 illustrates the variations of the concentration of CO₂, O₂, dilution ratio and temperature of the flue gases during the combustion cycles obtained from all the batches of the 20 kW wood stove. In Figure 2, it is observed that O₂ concentrations increase slowly and CO₂ concentrations decrease slowly during the combustion phases. This might be explained by the combustion of the volatile hydrocarbons starts with stable combustion temperature. It contains many complex chemical reactions, which are parallel and sequential. The combustion is mainly based on the thermal decomposition of cellulose and the reactions

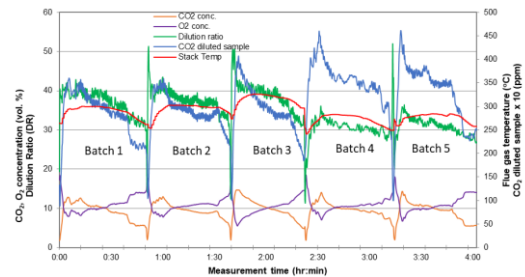


Figure 2. Variation of concentration of CO₂, O₂, dilution ratio and stack temperature obtained from all batches of 20 kW stove

of pyrolysis products with each other and with gases in the air, mainly oxygen. It can also be seen from the figure that the DR varies with the variation of CO₂ concentrations of the raw sample and diluted sample by applying dilution air flows into the PRD and ED from the measurements. the starting time of a new batch. The DR was quite stable during each measurement. The average DR values in different batches of the stoves varied between 26 to 45 for the startup phase, 31 to 39 for the combustion phase and 30 to 41 for the total cycle.

Figure 3 is the time series of particle number concentrations obtained from Batch 1 to Batch 4 of the 10 kW stove obtained from the nominal heat output. Batch 5 operated with part load heat output is not presented in the curve. It is observed from the figure that number concentrations varied significantly over time for the different batches, which is typical for small scale wood stove combustion. The figure shows the number concentrations sharply increased in the startup phase of the biomass combustion cycle. The startup phase is characterized by higher particle emissions than combustion phase with stable operation, whereas the burnout phase of the fuel conversion process consists mostly of char combustion and majority of the particle are bottom ash. The vertical blue dash lines indicate the starting time of a new batch.

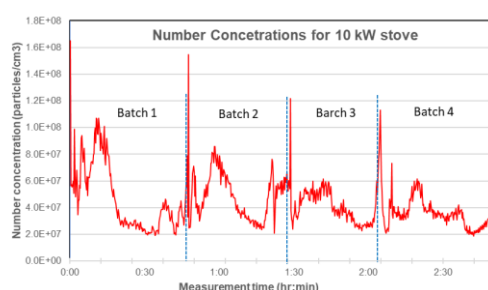


Figure 3. Time series of particle number concentrations obtained from the measurements of the 10 kW stove

Particle Number Concentrations

Generally, particle number concentration is defined as the number of particles per unit volume of flue gas. The unit expressed in number concentrations is particles/cm³. Figures 4 and 5 compares the particle number concentrations emitted from different phases of each batch of the 10 kW stove and the 20 kW stove respectively.

The total particle number emissions represents as the amount of particle concentrations from the startup to the end of the combustion phases. It can be seen from the Figures 4 and 5 that particle number concentrations measured from both stoves

varied between 3.4×10^7 to 8.7×10^7 particles/cm³ for the startup phase, 2.6×10^7 to 5.0×10^7 particles/cm³ for the combustion phase, 1.8×10^7 to 2.7×10^7 particles/cm³ for the burnout phase and 2.7×10^7 to 5.3×10^7 particles/cm³ for the total cycle. For the 10 kW stove, Batch 2 had the highest total particle number emissions than that in Batch 3 followed by Batch 1, Batch 4 and part load experiment Batch 5. Particle number emissions from the part load Batch 5 is close to the concentrations of the nominal load operated Batch 4. This is probably higher excess air factor ($\lambda = 2.14$) obtained in Batch 5 leading to lower combustion temperature (258°C). On the other hand, lower air excess factor ($\lambda = 1.87$) for the Batch 4 created better combustion conditions (flue gas temperature 292°C) leading to lower particle emission as PM emission is strongly dependent on the combustion conditions. While for the 20 kW stove, Batch 1 had the highest total particle emissions, followed by Batch 2, Batch 3, Batch 5, and Batch 4. In the Batch 4 of 10 kW stove the lowest particle number emissions were observed at 25% primary air setting while, in the Batch 4 of 20 kW stove at 15%.

Both figures show that particle number emissions in the startup phase were relatively much higher than those in the combustion phase. The elevated particle number formation in the startup

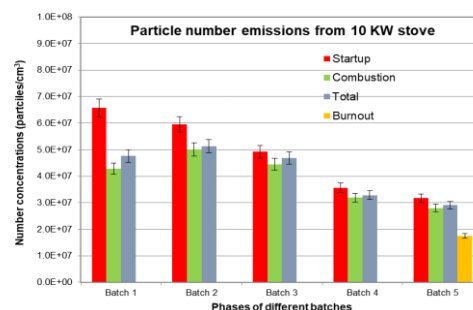


Figure 4. Particle number emissions from different phases of the 10 kW stove

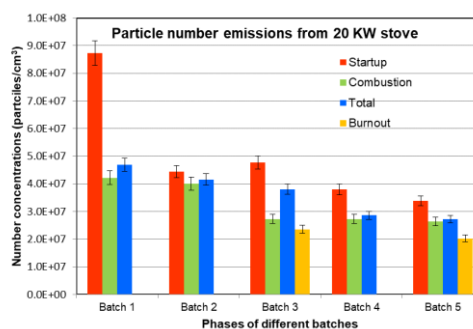


Figure 5. Particle number emissions from different phases of the 20 kW stove

phase can be explained by incomplete combustion as the initial combustion processes such as wood drying, pyrolysis, devolatilization, etc. also have an impact on the number emission levels. This is probably due to the large amount of volatile matter released from the wood log fuel during the startup phase. These species later saturate and form fine particles by nucleation. The nucleated smaller particles grow further by coagulation, agglomeration, condensation and surface reaction. Besides, the startup phases operated with low temperature condition than the main combustion phase. The difference of the average flue gas temperature between two phases is observed to be about 20°C for the 10 kW stove and 23°C for the 20 kW stove. Another reason for higher number concentration in the startup phase is might be dust carryover from the previous phases.

The duration of the startup phase (about 5-7 min) is much shorter than the main combustion phase (about 40-46 min), though the duration in the startup phase was very short but significant particle emissions produced during this phase and emitted directly to the atmosphere. As can be seen of the both figures, total particle number emissions from all the batches (except three batches: Batch 1 of 10 kW, Batch 1 and 3 of the 20 kW) of both stoves were more or less close to the emissions of the combustion phases. Therefore, it can be stated from our findings that the impact of the higher particle number emissions during startup on the total particle emissions is minor.

Particle number emissions from both stoves show that the highest particle number concentrations occur during the startup phase next the combustion phase followed by the burnout phase. The ash from the preceding batches of the combustion cycles and the preheating procedures was staying in the combustion chamber of the burnout phase. In the burnout phase, usually no combustible gases are present, only the char fraction burns and majority of the particles are bottom ash.

The particle number emissions from the different phases of both wood stoves differ from one batch to another, which are typical for the combustion of wood stoves. This may be due to the difference of combustion chamber volume, fuel consumption rate, primary air distribution and heat output. The particle formation and concentrations are strongly influenced on various aspects: fuel type and size, moisture content, chemical composition of the fuel, air preheating, primary/secondary air distributions, combustion temperature, residence time, turbulence, sufficient draft, full load and part load operation, excess air ratio, combustion technologies, etc. (Johansson *et al.*,

2003; Bari *et al.*, 2011; Obaidullah *et al.*, 2012a; 2012c; Obaidullah and De Ruyck, 2020).

The number concentrations results obtained in our study are similar the results obtained in other studies. For example Johansson *et al.* (Johansson *et al.*, 2003) investigated particle number concentrations from biomass combustion in a pellet burner of 6 kW using an ELPI and the experimental results show that number concentrations varied between 1.8×10^7 and 8.7×10^7 particles/cm³. Experimental results from our study are in agreement with Gaegauf *et al.* (Gaegauf *et al.*, 2001) who reported particle number concentrations from biomass combustion systems. Another study, Sippula O. (Sippula, 2010) examined fine particle emissions from a 8 kW pellet stove using and EPLI for particle emissions characteristics and observed that number concentrations varied between 1.1×10^7 and 4.7×10^7 particles/cm³.

Since small scale biomass combustion appliances are considered as an important source of particle emissions, therefore the regulations of the standard can be discussed. The existing European Union (EU)/US Environmental Protection Agency (EPA) norms limit the particle emissions in terms of total mass concentrations from the combustion appliances (Mudgal, 2009; Villeneuve *et al.*, 2012). However, nowadays much attention is focused across the globe as well as in the EU member states to extend this approach to the particle number concentrations and size distributions, because of their impact on the global environment, the air quality and the adverse human health (Obaidullah *et al.*, 2018c; 2019a; Obaidullah and De Ruyck, 2020).

Particle Number Size Distributions

Startup Phase

Particle number size distribution is defined as the number concentration of particles distributed over particle size. Figures 6 and 7 show that particle number size distributions obtained from the startup phase of different batches of the 10 kW stove and 20 kW stove respectively. The abscissa represents the particle aerodynamic diameter in logarithmic scale plotted against the ordinate which shows the ratio of number concentration dN to the logarithm of the channel width $d\log(Dp)$, where Dp is the aerodynamic diameter. Since, the formation mechanism of the particulate matter is quite complex and usually includes several concurrent paths, the particle number size distributions may reveal more than one peak. It can be seen from the Figure 6 for the 10 kW stove that three batches

(Batch 3, Batch 4, and Batch 5) have bimodal particle size distributions with a maximum particle number concentration at a particle size of about 20 nm in one mode while in another mode the concentration peak appears at the particle size of about 125 nm. Other two batches (Batch 1 and Batch 2) have the unimodal particle size distributions with maximum particle concentration at the particle size around 20 nm for Batch 2, while peak particle concentration appears at the particle size around 40 nm for Batch 1. The particles with diameter below 100 nm are the most important when considering the number distributions, but it contributes only a very small fraction of the total mass of particles.

It can be seen from the Figure 7 that all the batches show a quite similar distributions with two peaks (bimodal size distributions). One mode has a peak particle concentration at the particle size around 20 nm and another mode has a peak concentration between 70 nm and 125 nm. Batch 1 had the highest particle number concentration, which is similar to the 10 kW stove. From the particle number size distributions in the startup phase of the 10 kW and 20 kW wood stoves, it can be concluded that all the batches showed their maximum particle concentration in the ultrafine

region, which is considered very harmful for human health as they penetrate the alveolar region of the lung.

Combustion Phase

Figures 8 and 9 presents the curves of the particle number size distributions obtained from the combustion phase of all the batches of the 10 kW and 20 kW stove respectively. The Figure 8 shows that all the batches have bimodal particle size distributions for 10 kW stove. One mode has maximum particle concentration at particle size around 20 nm and another mode has a highest maximum at 125 nm for all batches. The particle size distributions of the 10 kW stove shows that the second mode (125 nm) has significantly higher particle concentrations than the first mode (20 nm). It can be seen from the particle number size distributions of the Figures 6 and 8 that the particle number concentrations in the first mode of the combustion phase are much lower than that in the first mode of the startup phase of the 10 kW stove. This is probably less soot particle emissions in the first mode and high fine fly ash particle emissions in the second mode of the combustion phase. Similar bimodal distribution curves of particle

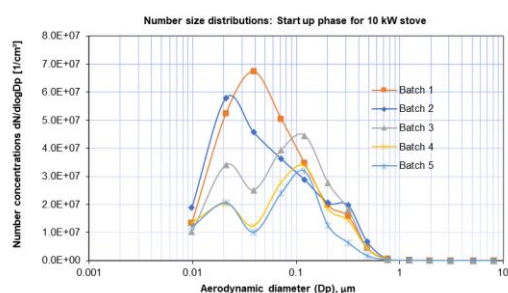


Figure 6. Particle number size distributions obtained from all batches during the startup phase of 10 kW stove

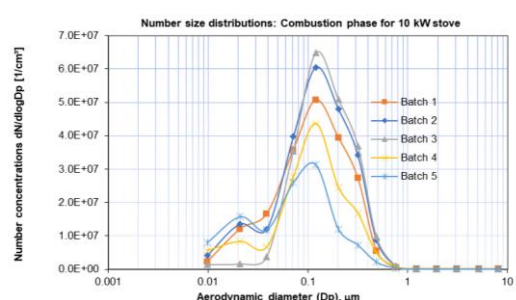


Figure 8. Particle number size distributions obtained from all batches during combustion phase of 10 kW stove

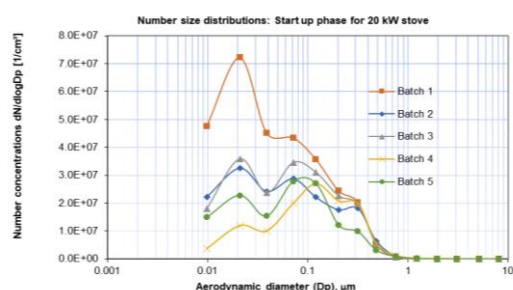


Figure 7. Particle number size distributions obtained from all batches during the startup phase of 20 kW stove

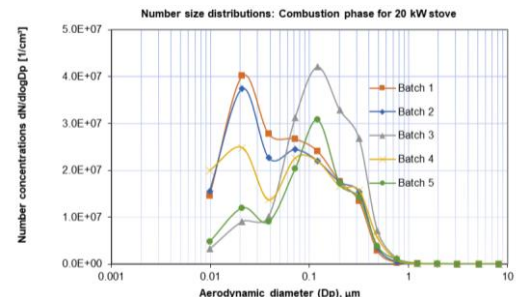


Figure 9. Particle number size distributions obtained from all batches during combustion phase of 20 kW stove

number emissions for 20 kW stove are presented in Figure 9. In this case, the number size distributions from the curve for all the batches show that maximum number concentration of particles at the size around 22 nm and 125 nm.

Similar particle number size distributions have appeared in other works. For example, Bari *et al.* (Bari *et al.*, 2011) examined particle number concentration using an SMPS in a wood fired heating appliance with a capacity of 5 kW. They found that maximum number concentrations of the emitted particles were unimodal in the range about 60 nm to 100 nm. Pettersson *et al.* (Pettersson *et al.*, 2011) investigated number size distributions from a 9 kW wood stove using an SMPS and they found that maximum number concentrations observed at the 100 nm to 300 nm for the combustion phase, which is relatively higher particle size from this study.

Burnout Phase

The ashes from the preheating process and previous batches that remained in the combustion chamber were burnt in the burnout phase. Figure 10 shows the particle number size distributions obtained from three measurements during the burnout phase of both stoves. Measurement 1 obtained from the Batch 5 of the 10 kW stove and Measurements 2 and 3 obtained from the Batch 3 and Batch 5 respectively of the 20 kW stove. It can be seen from Figure 10 that maximum number concentrations from the distribution curves are observed at two distinctive modes of particle size around 20 nm and 70 nm. The first mode 20 nm shows the higher particle number concentration than the second mode 70 nm. Similar results are observed by other studies studying various types of the wood stoves using an Electrical Low Pressure Impactors for the particle emission analysis (Bäfver *et al.*, 2011; Ozgen *et al.*, 2013). For example, Gaegauf *et al.* (Gaegauf *et al.*, 2001) used

a Scanning Mobility Particle Sizer (SMPS) to show that the number size distribution during the burnout phase is located at the particle size of about 30 nm, which is close to the results of the present study.

Number size distributions from all phases showed dominant quantities of ultra-fine particles. This is an important parameter because it affects the air quality much than particle mass. Particles numbers between 1 μm and 10 μm represent a very few number of particles (probably coarse fly ash particles), therefore these particles are not seen in the number size distribution curves. However, when considering mass size distributions, particles between these sizes (1 μm and 10 μm) show maximum concentration of particle mass.

It is observed from the number size distributions analysis that the lower impactor stages 1 to 9 of the ELPI+ dominate the ultrafine particle (diameter below 100 nm) concentrations, with the values of 99.91%, 99.81% and 99.94% of total number concentrations for the startup, combustion and burnout phase respectively. These particles represent the main part of the total number of particles emitted from both stoves. These ultrafine particles are probably soot and fly ash particles. Soot particles are complex mixture consisting mainly of amorphous elemental carbon and organic material. Soot formation starts during devolatilization and combustion of volatiles when hydrocarbon fragments leave from the fuel particles. These fragments then crack into smaller pieces and react with one another and surrounding gases to form aromatic rings. These aromatic rings then start to form polycyclic aromatic hydrocarbon (PAH). Further growth of PAH leads to form soot. Generally, two pathways for the formation of soot nuclei can be discussed. First, at lower temperatures, aromatic hydrocarbons produce soot directly by growing into graphite-like structure. Second, at higher temperatures both aliphatic and aromatic hydrocarbons first fragment, which are followed by polymerization of the fragments to larger molecules, forming soot. Thereafter, the particles grow larger by surface reactions, coagulation and agglomeration (Knudsen *et al.*, 2004; Wiinikka, 2005; Obernberger *et al.*, 2006; Sippula, 2010; Obaidullah *et al.*, 2012a; 2018c; 2019a).

The fine fly ash particles (diameter <1 μm) are formed from the easily volatile inorganic elements, released from the fuel to the gas phase during combustion. Potassium, sulphur and chlorine are the most relevant element during the combustion of biomass fuels whereas sodium and heavy metals (zinc and lead) provide minor contributions. In the gas phase, these species undergo several reactions resulting in the formation of alkaline (K, Na) metal chlorides, sulphates, carbonates, and heavy metals

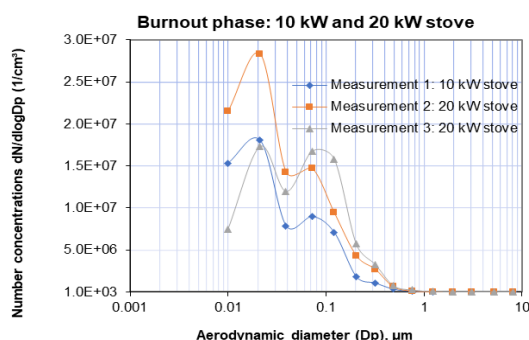


Figure 10. Particle number size distributions obtained from burnout phase of the 10 kW and 20 kW stove

oxides (Wiinikka, 2005; Obernberger *et al.*, 2006; 2007; Obaidullah *et al.*, 2012a; 2018c; 2019b). These fine ash particles are formed by nucleation, condensation, surface reactions, coagulation and agglomerations. The fine particles are mainly formed of potassium salts such as K_2SO_4 (potassium sulphate), KCl (potassium chloride), K_2CO_3 (potassium carbonate) and smaller amounts of zinc, sodium and organic material.

Conclusions

The experimental results presented in this paper are particle number emissions and size distributions from several batches of two commercially available Belgian made modern wood stoves. The combustion experiments were conducted in a laboratory of a stove manufacturing plant. Following conclusions can be drawn from this study.

Particle number concentrations in the flue gas obtained from both wood stoves experiences large variation during the different phases of the combustion cycle. Particle number concentrations measured from both stoves varied between 3.4×10^7 to 8.7×10^7 particles/cm³ for the startup phase, 2.6×10^7 to 5.0×10^7 particles/cm³ for the combustion phase, 1.8×10^7 to 2.7×10^7 particles/cm³ for the burnout phase and 2.7×10^7 to 5.3×10^7 particles/cm³ for the total cycle. The startup phase of all the batches was characterized by higher particle number emissions than in the combustion phase followed by the burnout phase. Higher particle concentration in the startup phase is probably due to lower stack temperature and dust carryover.

Particle number size distribution results show that all the phases have bimodal size distributions. One mode has a peak particle concentration at a particle size around 20 nm while another mode has maximum concentration between 70 nm to 125 nm. These ultrafine particles are very harmful for human health as they penetrate the alveolar region of the lung. This is an important parameter because it affects the air quality more than particle mass.

Since the fine particles are believed to be more harmful, more attention should be given to fine particle regulations. Since their mass is not so relevant, the regulations should focus on the particle amounts in number rather than mass.

Since small scale biomass combustion appliances are identified as an important source to local PM emissions, there is an increasing interest for particle removal from these appliances. In order to reduce particle emissions from these appliances, flue gas cleaning technologies can be used. Major flue gas cleaning systems applied in the stack of the residential biomass combustion appliances are

cyclones, electrostatic precipitators (ESPs) and wet scrubbers. ESP systems have higher particle separation efficiency than the cyclones and wet scrubbers and use in the EU countries. However, such particle removal technologies are quite expensive for small scale applications.

Particle emissions can be reduced by installing ESP units in the stack. Chemical analysis on our collected PM was not performed. More information is needed on the role of the chemical composition and physical structure of fine particles on adverse health effects and climate effects. Therefore, a logical next step would be the determination the chemical content of the fine particle emissions.

Acknowledgments

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