EMISSION FACTORS OF GASEOUS POLLUTANTS FROM SMALL SCALE COMBUSTION OF BIOFUELS

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ABSTRACT

The paper presents results of determination of emission factors for CO, NO_x and SO_2 from combustion of wooden and various sorts of agricultural biofuels in a commercially available small scale pellet boiler. The emission factors (EF) were determined at three different power loads of the boiler and are presented in mass, LHV and utilizable energy form. The results show significant influence of operating conditions as well as conditions of the boiler itself. Comparison of the results with various literature data shown significant impact of the experimental procedure on CO EFs and turned out that definition of the experimental conditions is essential for relevance of the presented data. EFs for NO_x and SO_2 were found to be particularly affected by nitrogen and sulphur content in the biomass respectively.

KEYWORDS

biomass; combustion; emission factor; small scale; boiler

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1. INTRODUCTION

Small scale residential biomass combustion has been proved to significantly contribute to local quality of air. Therefore, it is important to be able to evaluate amounts of pollutants released into the atmosphere from these individual sources (Williams, et al., 2012). The emission limits for appliances above 500 kW of the power capacity are established and measurement of concentration of pollutants must be done either periodically or continuously. The Directive 2010/75/EU is the general document of the EU concerning the emission limits from stationary combustion sources and it sets the emissions limits for nominal power load capacities above 50 MW (Official Journal of the European Union, 2010). The power capacity range from 0.5 to 50 MW is currently covered by national legislations. The governing document for small scale combustion below 500 kW is the standard EN 303-5. This standard was introduced in 1999 and it was updated in 2012 (European Committee for Standardization, 2012). The standard classifies the small scale boilers into the emission classes. The version EN 303-5:1999 defines emission classes 1-3, while in the updated EN 303-5:2012 are added the classes 4 and 5 and the classes 1 and 2 are removed.

The emission limits for NO_x are not included in the EN 303-5 standard. They are currently included only in national legislation of Austria (Bundeskanzleramt Rechtsinformationssystem, 2011) for small scale boilers. The emission limits are set to 150 mg/MJ for wooden pellets and logwood (100 mg/MJ from 2015) and 300 mg/MJ for non-wooden standardized biofuels in automated feeding boilers. The standard EN 303-5 describes testing procedure that must be carried out for each type of the boiler before introducing on the market. Testing of the boiler is performed at nominal capacity load and lowered load at 30 % of the nominal value while the limits given by the EN 303-5 must be met for the nominal capacity load. According to the 2012 update, the limits for PM must be met for both testing loads.

When the boiler comes to the market after the testing procedure, no further measurement of emissions is required during its operation. An exception is Germany where a check of the CO and PM limits must be made every second year (Bundesministerium der Justiz, 2010). It is obvious that real levels of the emitted pollutants can be totally different from laboratory testing. The real emissions mostly depend on quality of fuel, way of operating the boiler and maintenance of the boiler. Therefore, it is difficult to make a reliable calculation of amount of the emitted pollutants in a certain urban area without knowing the real emission levels.

Using the emission factor is a way how to evaluate these levels of pollutants emitted from a combustion source if a direct measurement cannot be carried out, typically for domestic small scale boilers. Such a measurement is technically and legally complicated and also expensive. The emission factor is typically defined as the amount of a concerned pollutant emitted per the unit of burned fuel mass or per a defined task performed. This is often referred to the mass-based emission factor and has units such as g of a pollutant per kg of burned fuel. An alternative representation is done by the amount of a pollutant per MJ of calorific value of the fuel. Emission factors can be alternatively defined as the amount of emitted pollutant per some defined task performed. They can be called task-based emission factors (Mitra, et al., 2002), (Bhattacharya, et al., 2002) and can be expressed in different ways, e.g. amount of a pollutant related to a kWh of produced energy or to mass unit of a final product. When an emission factor is presented, it should be always accompanied by description of the testing procedure and by the conditions how was it obtained. If such an information is missing, the value of the emission factor cannot be considered as reliable.

In the literature, there can be found several studies that were aimed on measurement of the emissions factors from small scale biomass combustion, but results are quite different and there are only few references that involve features of operation of an automated domestic biomass boiler. If such a work is published, then it typically uses wooden pellets as the tested fuel. However, current trends in domestic heating show increase in using non-wooden biomass, typically agricultural residues and energy crops. A short overview of the published work is presented here and some of the interesting results are used for comparison in the Result and discussion section. Authors (Bhattacharya, et al., 2002) studied emission factors from wood and charcoal fired cookstoves that were manually fed by the fuel and obtained emission factors in mg/kg of fuel and g/MJ of useful heat energy. The publication by (Ndiema, et al., 1998) was also focused on biomass stoves. In both references, biomass stoves, that are recently used only rarely for domestic heating, were used (van Loo, et al., 2008) and therefore do not have significant relevance for European conditions. Authors (Ortiz de Zárate, et al., 2000) studied emission factors from combustion of cereal residues, but the experimental work was carried out in a testing chamber in order to simulate an open fire, which does not correspond to the conditions in residential heating systems. The work by (Wiinikka, et al., 2004) concerned a small scale pellet combustor and was focused particularly on emission of particulate matter. In addition, the authors evaluated emission factors for CO and NO_x given in mg per MJ of calorific value of the fuel. Wooden pellets were used in this case. The work published by Chinese authors (Cao, et al., 2008) is very interesting by studying emission factors from combustion of various crop residues. However, the work was aimed at simulation of combustion in stoves for cooking in rural areas. In the work by (Yuntenwi, et al., 2008) there were studied emission factors of CO and PM from biomass cook stoves. An extensive work was done with standardized pellets in both manual and automatically fed appliances with both wooden and non-wooden materials by (Schmidl, et al., 2011), however, this work was focused particularly on PM emissions and measurement of gaseous pollutants was not converted to emission factors by the authors. A study focused on emissions from various kinds of pellet boilers in real operation regime was done by (Win, et al., 2012). All boilers used in this study were market available and soft-wood pellets were used for testing. The test comprised of start-up and stop phase with six days of operation between. However, no more information is provided concerning the condition of the boilers. The work by (Johansson, et al., 2004) was also focused on pellet-fired boilers, where the authors compared modern and old-type boilers, again with wooden pellets. Probably the most recent and the most comprehensive work concerning several appliances and combustion cycles was done by (Ozgen, et al., 2014), however using only wooden biomass. To the recent investigations also belongs work done by (Amaral, et al., 2014), but this work was focused particularly on Amazon hard- and soft-wood batch combustion characteristic. Emission factors from small scale combustion are also published in the EEA emission inventory guidebook (Trozzi, 2009). It is a comprehensive and for public available overview, but it lacks details about conditions at which the emission factors were obtained.

The work presented in this paper used a commercially available automatically fed 25 kW pellet boiler. Combustion tests were carried out for four different materials, including wood and non-wooden pellets, at three different power loads of the boiler. This approach aims at reflecting real operation conditions of a small scale boiler during the heating period. The emission factors were obtained for CO, NO_x and SO_2 related to mass unit of fuel, MJ of calorific value of the fuel and MJ of useful heat energy.

2. MATERIALS AND METHODS

2.1 Fuels and experimental set-up

Five different sorts of biomass fuels were chosen for evaluation of the emission factors – wood, rape straw and grain straw in form of pellets, jatropha residues in form of pressings and palm nut shells. Selection of the biomass sorts covers traditional wooden biomass and residues from agricultural products. The residues from jatropha were chosen due to the fact that it is a potential top candidate plant for biodiesel production and the residues can be energy recovered (Barta, 2007). The palm shells are residual material from production of palm oil. Some results of combustion experiments using the shells as biological fuel are reported in literature, e.g. by authors (Ninduangdee, et al., 2014). Relevance of these residues is its production in high quantities with only a little utilization and currently is under consideration for co-firing with coal in CHP plants (Kavalek, et al., 2013). The pellets were standardized having 8 mm in diameter and in average 3 cm in length. Dimensions of the jatropha pressings were in the form of 3 mm thin layers, crushed into approximately 2 x 2 cm parts. Palm shells had dimensions in average about 0.5 x 1 x 0.5 cm. Proximate and ultimate analysis of the fuels is summarized in the table 1.

Table 1: Proximate and ultimate analysis of the biomass fuels

	Wood	Rape plant straw	Grain straw	Jatropha residues	Palm shells
as received					
LHV [MJ/kg]	16.35	15.13	17.60	17.56	17.35
water [wt. %]	7.8	9.3	6.4	8.0	7.5
ash [wt. %]	1.5	8.4	6.3	5.5	3.3
dry ash free					
C [wt. %]	51	50.4	49.3	44.2	51.7
H [wt. %]	6.9	7.1	7.5	5.4	7.0
N [wt. %]	0.3	1	0.8	0.1	0.4
S [wt. %]	0.003	0.06	0.1	0.07	0

A commercially available grate boiler for pellets with automated front fuel feeding was used for combustion experiments. The nominal rated capacity is 25 kW for wood and 18.5 kW for agricultural pellets. The main difference compared to the data obtained from the literature sources is that the boiler had already been in operation for around seven years that can be considered as satisfactorily long time to reflect the real-life operating conditions. This helps to make the results more representative and avoids unwanted improvements of results that may be caused by laboratory conditions. The boiler undergoes only irregular maintenance in the form of manual cleaning of the heat exchanger section. The boiler was connected to a controlled air cooled heat exchanger in order to keep water temperature difference at values typical for domestic heating system 80/60°C. Scheme of the experimental set-up is shown in figure 1.

The experimental procedure was based on carrying out combustion tests with the fuels at three power load levels. At first, all fuels were tested at 18.5 kW that is the nominal load for agricultural fuels. The wooden pellets were tested at this load level as well in order to be able to compare them with other fuels. Other loads were 12.5 kW and 7.5 kW, representing 67 and 40 % of the nominal load. Each single experiment lasted for two hours at stable conditions. During the experiments concentrations of flue gas components O_2, CO_2, CO, NO_x and SO_2 at the sampling point at the outlet from the boiler were continuously measured. Other recorded parameters were the fuel consumption by balance weighting, flowrate of water through the boiler water system and temperature difference between outlet and inlet. These data were used for evaluation of balance of the boiler. The required power load was controlled by adjustment of fuel feeding and amount of combustion air was controlled according to the control curve in figure 2 that was measured for this specific boiler by authors (Hrdlicka, et al., 2011). The amount of combustion air is determined by the excess air ratio λ in the figure 2.

Figure 1: Scheme of the experimental pellet boiler

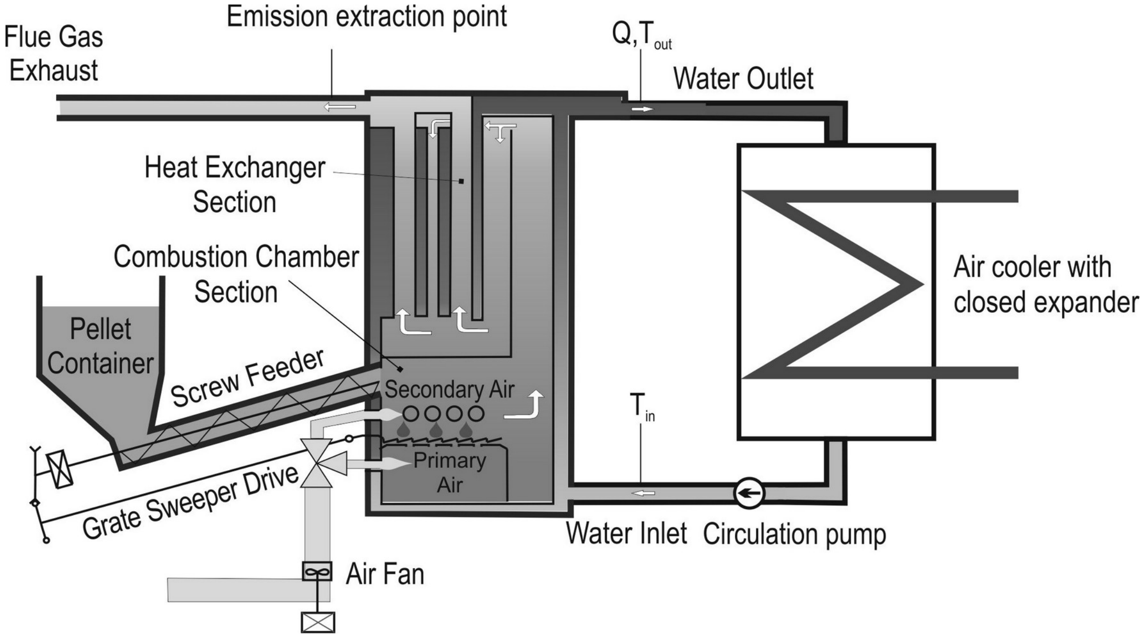
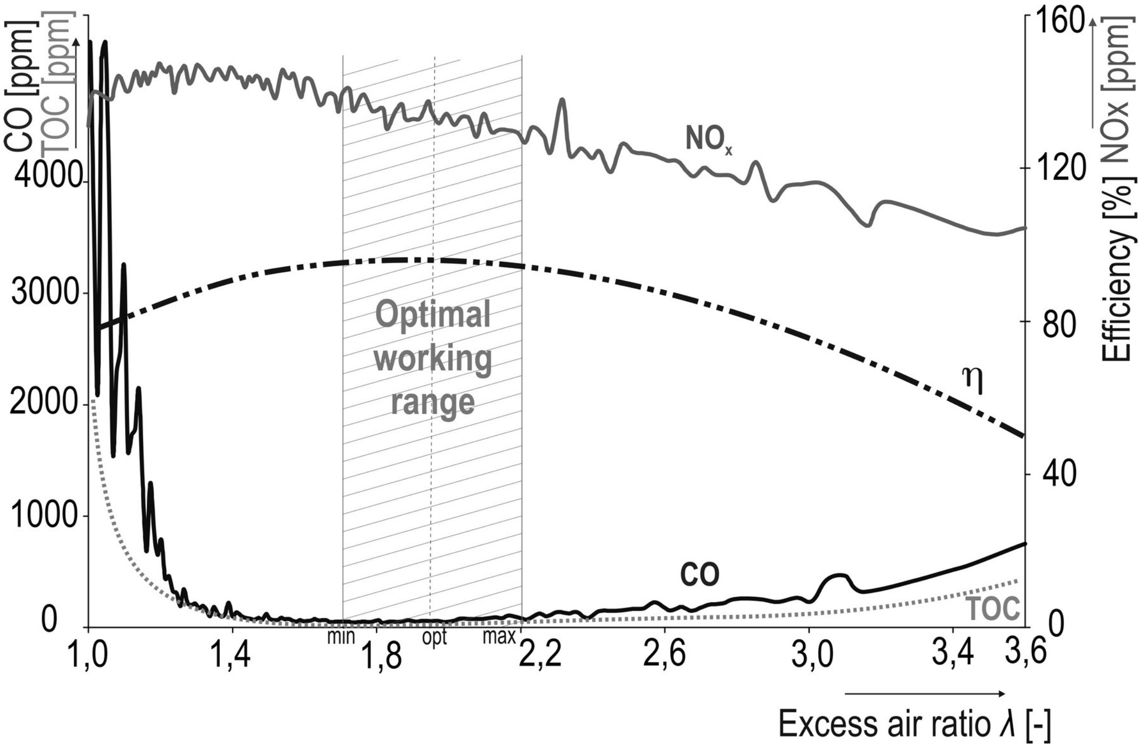


Figure 2: Control diagram of the pellet boiler for setting of combustion air (Hrdlicka, et al., 2011)



2.2 Calculation of the emission factors

The first step to obtain the emission factors of the gaseous components in the flue gas was recalculation of the measured concentrations from volume to mass fraction, using the equation:

$$C_m^X = C_V^X \cdot \frac{M_X \cdot p_{ref}}{R \cdot T_{ref}} \cdot \frac{(21 - O_{2ref})}{(21 - O_{2meas})} \quad [\mathrm{mg/m^3_N}] \qquad (1)$$

O_{2ref} was set to 10 % according to the standard EN 303-5. The second step is calculating the real specific volume of the combustion air and the flue gas based on contents of C, H, N, S and O elements in the fuel, using following equations:

$$V_{CA} = [1.864C + 5{,}554H + 0.698S - 0.700O] \frac{1}{0.21} \quad [\mathrm{m^3_N/kg}] \qquad (2)$$

$$V_{FG} = 1.855C + 0.0003V_{CA}\lambda + 0.800N + 0.7805V_{CA}\lambda + 0.0091V_{CA}\lambda + 0.21(\lambda - 1)V_{CA} \quad [\mathrm{m^3_N/kg}] \qquad (3)$$

In the equations 2 and 3 there are used data from the Table 1 in form of mass fractions of the respective elements that are recalculated to “as received” basis. The equations 2 and 3 assume complete combustion, which means that all carbon is oxidized to CO_2 and no unburned carbon in form of CO or char remains. If this assumption is not satisfied, certain error can be introduced in the calculation. According to the experimental experience of the authors this error does not exceed 5 % relative under typical combustion conditions. These typical conditions are approximately below 5000 ppm of CO concentration and below 15 % of unburned char in ash. Content of oxygen in the fuel is calculated as balance to 100 %. The value of excess air ratio λ is calculated from measurement of oxygen concentration in the flue gas:

$$\lambda = \frac{21}{21 - O_{2meas}} \quad [-] \qquad (4)$$

Since the specific volume of flue gas V_{FG} is related to a unit of the combusted fuel, the emission factor for each flue gas component related to a unit of the fuel can be obtained by equation:

$$EF_{mass}^X = V_{FG} C_m^X \quad [\mathrm{mg/kg}] \qquad (5)$$

The emission factor related to the LHV of a fuel is then obtained as follows:

$$EF_{LHV}^X = \frac{EF_{mass}^X}{LHV} \quad [\mathrm{g/GJ}] \qquad (6)$$

In the equation 6 the LHV is given in MJ/kg. The emission factor related to a unit of produced heat energy is based on following equation:

$$EF_{PE}^{X} = \frac{EF_{mass}^{X}}{\eta_B LHV} \quad [\mathrm{g/GJ}] \qquad (7)$$

The boiler's efficiency η_B is calculated by a direct balance of power output in heated water and power input in the fuel. An average value is taken over whole period of the experiment.

3. RESULTS AND DISCUSSION

3.1 Measured emission factors

Obtained emission factors are presented in following figures 3 – 5. Presented is mean value over the whole measurement period. The explanation of the acronyms is following:
W = wood
RS = rape plant straw
GS = grain straw
J = jatropha
PS = palm shells

Figure 3: Emission factors related to mass of combusted fuel at 100, 67 and 40 % of nominal capacity load

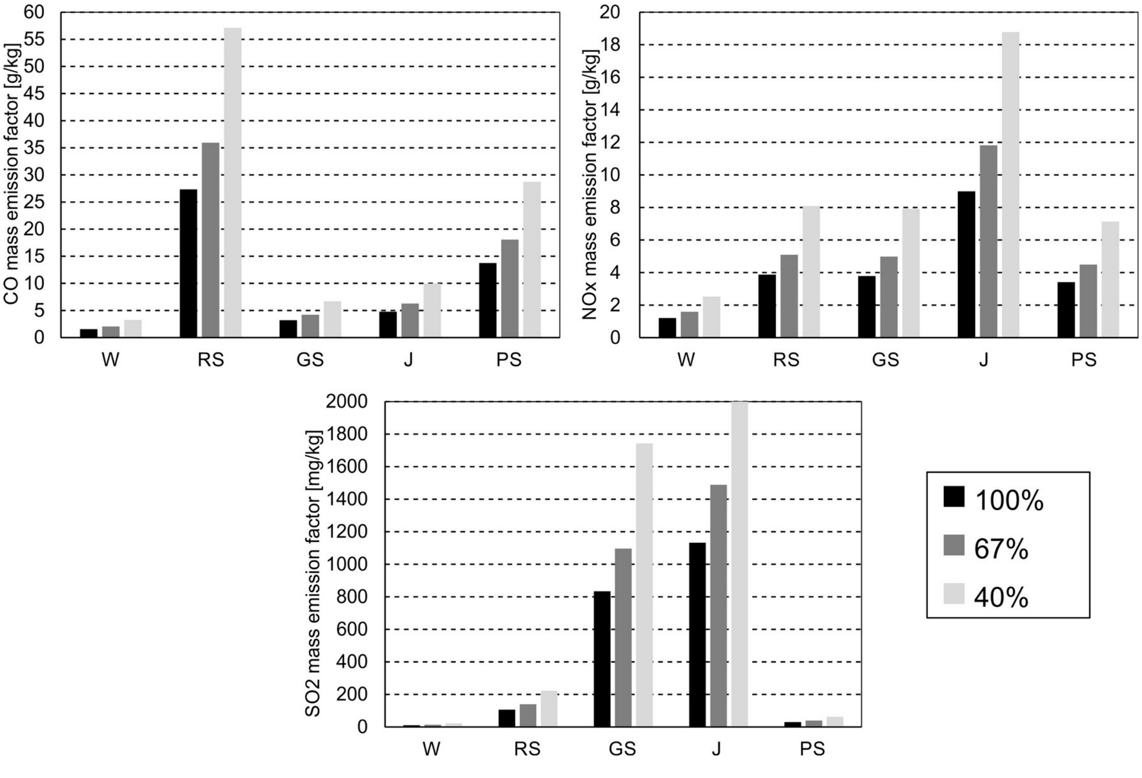


Figure 4: Emission factors related to LHV of a fuel at 100, 67 and 40 % of nominal capacity load

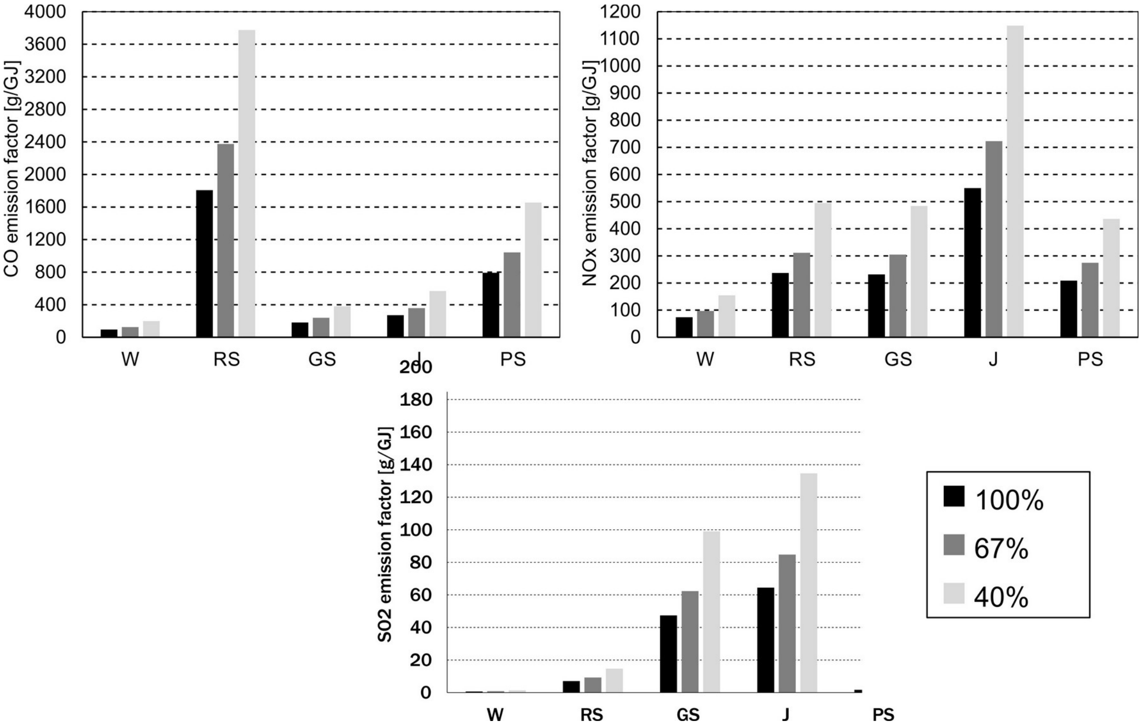
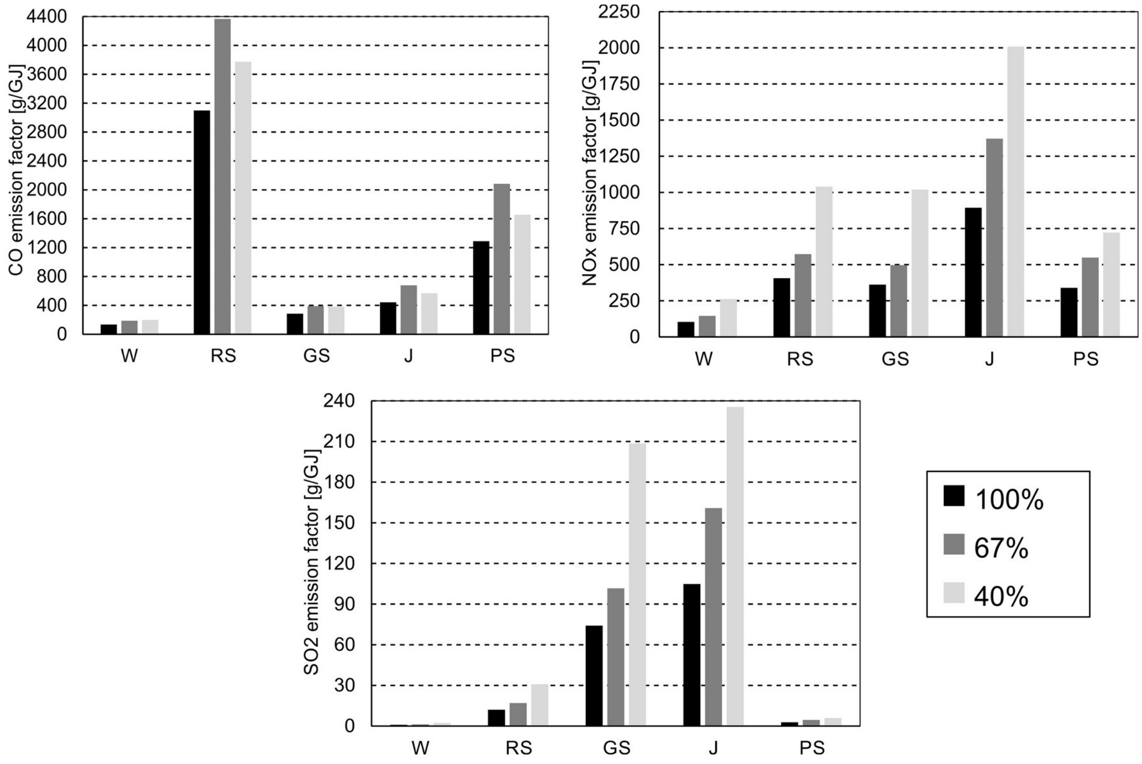


Figure 5: Emission factors related to GJ of useful energy at 100, 67 and 40 % of nominal capacity load



The emission factors related to the mass of burned fuel and LHV of the fuel are strongly influenced by power load level of the boiler. It can be clearly seen that with decreased power load of the boiler the emission factor rises. This effect is generally caused by a drop of thermal efficiency of the boiler at reduced power load. Generally for all measured gaseous pollutants, this drop is particularly caused by an increase of the excess air ratio, which increases the stack loss of the boiler. However, further differences can be observed among the gaseous pollutants. In case of the CO significantly higher concentrations at lower power load capacities were measured. These higher CO concentrations are typical result of decreased temperature in the combustion chamber caused by decreased load. The generally higher CO levels in the case of the agricultural crops combustion may also by affected by ash fusion, as reported by (Skopec, et al., 2014). The situation for NO_x emission factors is quite different. An increase of the excess air ratio at lowered load plays also some role, but the NO_x concentration does not change significantly with changing either power load or excess air ratio. The NO_x levels are particularly determined by nitrogen content in the fuel. Therefore the differences in NO_x emission factor among the levels of power load are not so high as for the case of CO. A special case is determination of the emission factors for SO_2. Biomass generally contains low amounts of combustible sulphur which results even in order of magnitude lower emission factors compared to CO and NO_x emission factors. Higher sulphur content can be found in the agricultural fuels (rape plant and grain straw and jatropha) corresponding with higher emission factors of SO_2.

The emission factors related to useful energy produced by the boiler are significantly affected by thermal efficiency of the boiler. The excess air ratio was the major affecting factor in the previous two cases. In this case is also important the outlet flue gas temperature from the heat exchanger system. This temperature affects together with the excess air ratio the stack loss of the boiler. As the result the differences in emission factors between the respective power loads are smaller than in the previous cases. The emission factor related to useful energy provides also the best representation of real life operation of the boiler, since it includes boiler's efficiency that does not depend on the combustion process only, but in the form of outlet flue gas temperature reflects heat transfer from the flue gas to the heating water.

3.2 Comparison with literature data

Comparison of the measured emission factors with literature data is complicated by different representation of the emission factors and often missing details about the experimental procedure. Only those references where their authors provided enough data for comparison, regardless of the fuel form and combustion device, were selected for comparison. Comparison of the mass emission factors is summarized in the table 2.

Table 2: Comparison of the mass emission factors in g of pollutant per kg of burned fuel as received

	APPLIANCE	FUEL	CONDITIONS	EF CO	EF NO_x	EF SO_2
THIS WORK	commercial 25 kW grate pellet boiler	wood	nominal load, efficiency 58-71%, 8-10.5 % O_2 in flue gas	1.6	1.2	0.01
		rape plant str.		27.3	3.9	0.1
		grain straw		3.2	3.8	0.8
		jatropha		4.8	9.0	1.1
		palm shells		13.8	3.4	0.03
SOURCE	APPLIANCE	FUEL	CONDITIONS	EF CO	EF NO_x	EF SO_2
Bhattacharya et al., 2002	simple rural area in Asia stoves. average	wood logs	efficiency at approx. 20 %	42	0.12	----
Ndiema et al., 1998	scaled down laboratory stoves	wood chips	6-12 % O_2 in flue gas	17.5	0.19	----
Ortiz de Zárate et al., 2000	laboratory testing chamber	cereal residues. without detailed specification	flaming and smouldering phase. without further details	---	2.8 dry matter	---
Cao et al., 2008	simple testing chamber	wheat straw	batch combustion, no other details	57.8	2.3	0.04
		rice straw		68	3.4	0.18
		corn stover		67.6	3.6	0.04
Yuntenwi et al., 2008	cooking stoves	wood logs. 15 % moisture	batch combustion, no other details	22	---	---
Ozgen et al., 2014	25 kW pellet boiler	wood pellets	averaged, nominal power load	6.6 dry matter	1.3 dry matter	---

The best comparable measurements are highlighted in the table 2 – wooden pellets at nominal load in a comparable appliance. It can be seen that the emission factors are very similar. When comparing the other fuels, it can be clearly observed effect of the appliance and testing procedure on CO emission factors. In principle, the simpler and less controlled appliance, the higher are the CO EFs. To the higher CO emission factors also contribute batch combustion that is typical for manually fed appliances. However, well comparable among completely different appliances are the NO_x emission factors. As can be confirmed from the table 2, their level does not much depend on appliance type or quality of the combustion process, but the determining factor is nitrogen content in the biofuel, which is also confirmed by results by (Cao, et al., 2008) who used agricultural products for the experiments. The SO_2 emission factor fully depends on content of sulphur in the biofuels and is not significantly influenced by choice of an appliance or way of its operation. They are generally very low and it is typical that the agricultural fuels show higher content of combustible sulphur compared to wooden based biofuels. Following table 3 shows results of this experimental work with authors that present data in the form of mass of pollutant per unit of LHV of a fuel.

Table 3: Emission factors in g of pollutant per 1 GJ of LHV of burned fuel as received

	APPLIANCE	FUEL	CONDITIONS	EF CO	EF NO_x	EF SO_2
THIS WORK	commercial 25 kW grate pellet boiler	wood	nominal load, efficiency 58-71%, 8-10.5 % O_2 in flue gas	95	74	0.7
		rape plant str.		1807	237	7
		grain straw		182	232	47
		jatropha		272	550	65
		palm shells		793	209	1.7
SOURCE	APPLIANCE	FUEL	CONDITIONS	EF CO	EF NO_x	EF SO_2
Wiinikka et al., 2004	10 kW laboratory pellet burner	6 mm wood pellets	9 % O_2 in flue gas	1.7	31.5	---
Ozgen et al., 2014	25 kW pellet boiler	low quality pellets	1 hour at nominal power load	594	129	---
		high quality pellets		107	13	---
Johansson et al., 2004	16 kW pellet burner	wooden pellets	11.7 % O_2 in flue gas. full load	30	---	---
			6.8 % O_2 in flue gas partial load 6 kW	380	62	---
Win et al., 2012	20 kW pellet boiler	wooden pellets	excluding start-stop phase, average	57	74	---
EEA emission inventory	boilers under 50 kW	wood and similar wooden waste	not specified	4000	120	30

The table 3 gives a good comparison of emission factors acquired by different authors using wooden pellets in appliances that are designed for combustion of the pellets. It is important to note that the CO emission factors for wooden pellets are in most of the cases at a comparable level. A significantly different value of EF for CO was obtained by (Wiinikka, et al., 2004) at about magnitude of order lower value. It is questionable whether this value is correct, since it would correspond at the specified conditions to concentration of the CO at about 1 mg/m^3_N in the flue gas. Such a low value is unlikely to be achieved in a solid fuel combustion appliance. The second significantly different value is presented in the EEA emission inventory. This value is about one up to two magnitudes of order higher compared to the rest of the emission factors. Nevertheless, in the EEA emission inventory is not specified the measurement procedure or data source of the emission factors. When comparing the agricultural fuels to the wooden biomass, it is evident that the emission factors for CO significantly rise. Comparison of the NO_x emission factors gives very similar results through all authors, particularly due to the fact that it is mainly given by nitrogen content in the biomass and there is no strong influence of combustion appliance or measurement procedure. The NO_x EFs for agricultural fuels are generally higher compared to wood, however no EFs for NO_x for other fuels than wood were reported by other authors in units of mass per calorific value. Neither SO_2 emission factors were reported by other authors in this form. The EEA emission inventory presents a value that seems to be an average over wider range of different sorts of biofuels. Interesting results also gives comparison of NO_x EFs with Austrian legislation, as described in the Introduction section. The upper limit for NO_x production is 300 g/GJ that was exceeded in case of jatropha and there is not much left to the limit for the straw-based pellets. It is a sign that it is likely that some sorts of agricultural biofuels might not be satisfactory considering the nitrogen content.

CONCLUSIONS

The determination of the EFs from a small scale pellet boiler in real conditions shown that operation conditions of the boiler must be always presented, typically power load and excess air ratio, otherwise the EFs are not meaningful. EFs in form of mass per unit of usable energy can be considered as the most representative since it considers thermal efficiency of the boiler that affects the emission factors significantly. Literature data are mostly consistent in EFs for NO_x and SO_2, but CO data significantly differ due to the choice of appliance and the way of the experimental procedure.

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LIST OF SYMBOLS

C	carbon mass fraction in the fuel [kg/kg]
C_m^X	mass concentration of the component “x” [mg/m^3_N]
C_V^X	volumetric concentration of the component “x” [ppm]
EF^X_{mass}	mass emission factor for the component “x” [mg/kg]
EF^X_{LHV}	energy in fuel related emission factor for the component “x” [g/GJ]
EF^X_{PE}	produced energy related emission factor for the component “x” [g/GJ]
H	hydrogen mass fraction in the fuel [kg/kg]
LHV	lower calorific value [MJ/kg]
M_X	molecular weight of the component “x” [g/mol]
N	nitrogen mass fraction in the fuel [kg/kg]
O	oxygen mass fraction in the fuel [kg/kg]
O_{2ref}	oxygen reference concentration [% vol.]
O_{2meas}	measured oxygen concentration [% vol.]

p_{ref}	reference (normal) pressure [kPa]
R	universal gas constant [J/(mol.K)]
S	sulphur mass fraction in the fuel [kg/kg]
T_{ref}	reference (normal) temperature [K]
V_{CA}	specific volume of combustion air [m^3_N/kg]
V_{FG}	specific volume of flue gas [m^3_N/kg]
η_B	boiler thermal efficiency [-]
λ	excess air ratio [-]

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