ABSTRACT: In this paper results from particulate emission measurements performed within a project at the Austrian Bioenergy Centre and the Institute for Resource Efficient and Sustainable Systems, Graz University of Technology, are summarised. Test runs at a test stand as well as field measurements with different modern Austrian small-scale pellet and wood chip furnaces as well as a log wood furnace (only boilers, no stoves were investigated) have been performed. The particulate emissions were characterised by their concentrations in the flue gas, particle size distributions and chemical compositions. The results clearly indicate that compared with older systems the particulate emissions of modern biomass furnaces are significantly lower. While the average fine particulate emissions of softwood pellet fired furnaces are in a range <20 mg/Nm³, emissions of wood chip fired furnaces are up to about 60 mg/Nm³ due to the utilisation of hardwood/softwood/bark mixtures. The log wood boiler investigated showed at stable load operation conditions the lowest emissions with <10 mg/Nm³. It has also been shown, that at insufficient gas phase burnout conditions, the formation of aerosols from unburned hydrocarbons and soot significantly increases the particulate emissions and therefore, a good performance of the process control system, a correct furnace design and, in the case of log wood boilers, the application of heat storage tanks in order to smoothen rapid load changes are of great relevance to keep particulate emissions low.

Keywords: combustion, aerosols, emissions

1 INTRODUCTION

In many European regions wood combustion provides a significant contribution to the domestic heating sector. Pellet furnaces, wood chip furnaces and logwood boilers represent today’s most important residential biomass combustion technologies. Due to increasing costs for fossil fuels as well as due to CO₂ mitigation reasons, the market for these applications has been rapidly expanding during the last years and this trend is expected to continue.

In parallel with the rising interest in biomass-based residential heating systems also R&D efforts increased with the aim to enhance the user friendliness of the applications, to increase efficiencies and to significantly decrease emissions. The success of this development work is documented by the trends of CO emissions and dust emissions determined during test stand measurements with Austrian pellet boilers in the last years presented in Fig. 1 and Fig. 2.

Figure 1: Development of CO emissions of new Austrian pellet furnaces

Figure 2: Development of dust emissions of new Austrian pellet furnaces

Therefore, it can be stated, that present state-of-the-art small-scale biomass combustion systems have already achieved comparably low levels of gaseous emissions. However, concerning particulate emissions, especially fine particulate emissions, still a disadvantage compared to oil and natural gas fired systems exists.

With the introduction of the EU directive 1999/30/EC, which limits PM₁₀ concentrations in the ambient air, it had to be recognised that in many European regions these limiting values are frequently exceeded. As the main sources for PM emissions traffic, industry and domestic heating have been identified. According to the Austria's informative inventory report (IIR) 2006 [3], in Austria the contribution of wood combustion to the PM emissions from domestic heating accounted for 88% in 2004.

Therefore, the public debate concerning particulate emissions automatically led to the discussion of particulate emissions from biomass combustion. In the course of these discussions it had to be recognised that almost no reliable data about PM₁₀ emissions from small-
scale biomass furnaces were available. Consequently, actual air pollution inventories are also based on old emission data which do not consider the rapid technological development and improvement of small-scale biomass combustion systems and thus the gradual reduction of particulate emissions by the substitution of old boilers by new boilers. Therefore, appropriate research activities focusing on the determination and characterisation of fine particulate emissions from small-scale biomass combustion systems have been initiated in many European countries and are still ongoing [4, 5, 6, 7]. One of these projects, specially related to the determination of fine particulate emissions from Austrian state-of-the-art small-scale biomass furnaces is performed at the Institute for Resource Efficient and Sustainable Systems, Graz University of Technology, in cooperation with the Austrian Bioenergy Centre (ABC), Graz. The main aim of this project is to characterise fine particulate emissions from small-scale biomass furnaces by their concentration in the flue gas, particle size distribution as well as chemical composition. Furthermore, the governing parameters influencing fine particle formation should be identified and strategies for emission reduction mainly focusing on primary measures should be worked out. The project focuses on all relevant small-scale residential biomass combustion technologies including modern automatic furnaces for pellets, wood chips and wood logs. Moreover, for comparison, small-scale oil boilers are tested too.

In this paper the results of the investigations performed with state-of-the-art automated small-scale pellet and wood chips boilers as well as with a log wood boiler are presented.

2 THEORETICAL BACKGROUND

2.1 Definitions

Ambient particulate matter (PM) is usually categorised in coarse mode (>1 µm) and fine mode (<1 µm) particles. Other classifications applied are TSP (total suspended particulate matter) which represents all particles present as well as PM_{10}, PM_{2.5} and PM_{1}, which involve the particle fractions with a diameter <10, 2.5 and 1 µm respectively.

These particle classes are always related to the aerodynamic particle diameter (a.e.d.) which represents the diameter of a spherical particle with a density of 1 g/cm³, showing the same sedimentation behaviour in air as the particle of interest. The introduction of the aerodynamic diameter provides the possibility to compare particles with different structures, shapes and densities. This is especially of relevance for ambient PM due to their high heterogeneity caused by their different particle sources and compositions.

Concerning particulate emissions from biomass combustion it can also be distinguished between a coarse mode (coarse fly ashes) and a fine mode (aerosols) (see section 2.3). Both together form the total fly ash emissions.

2.2 PM sources

Coarse mode particles mainly originate from mechanical processes such as abrasion and particle dispersion caused by e.g. traffic, agricultural and industrial processes. Fine mode particles are almost exclusively due to emissions of combustion or thermal processes.

Fine mode particles are mainly precipitated by fog formation and rainfall. Therefore, depending on the meteorological constraints, they show long residence times in the atmosphere. Consequently, also transboundary long distance transport of PM can impact a regional or local ambient air quality situation. Especially these transport phenomena make the allocation of PM emissions regarding a specific ambient air quality situation difficult.

2.3 Particle formation during biomass combustion

Compared to heating oil and natural gas solid fuels such as biomass and coal contain considerable amounts of ash forming elements. It must be distinguished between elements, which are non-volatile under the constraints of biomass combustion (Ca, Si, Mg, Fe, Al) and easily volatile elements like K, Na, S, Cl and easily volatile heavy metals (e.g.: Zn, Pb). While easily volatile elements are partly released to the gas phase (evaporated) during the combustion process, non-volatile elements remain in the solid phase.

The major share of ashes formed during fixed-bed biomass combustion leaves the furnace as so-called bottom or grate ash. This fraction mainly consists of refractory species (Ca, Si, Mg, Fe, Al) but also contains the not released part of the easily volatile elements. Moreover, a minor portion of ash particles is entrained from the fuel bed with the flue gas and forms the coarse fly ashes. Depending on the flow conditions coarse fly ash particles are partly precipitated in the furnace and the boiler. The particles remaining in the flue gas leave the boiler as coarse fly ash emissions. These coarse fly ash emissions are usually in a particle size range between some µm up to about 200 µm [8]. According to their formation mechanism, coarse fly ash emissions increase with increasing ash content of the fuel, increasing disturbance of the fuel bed and increasing flue gas velocities (increasing load of the combustion unit). Due to their rather big particle size their contribution to the PM_{10} emissions is usually of minor relevance.

The second type of particulate emissions formed during biomass combustion, which is highly relevant for the PM_{10} fraction, are fine particulate emissions, the so-called aerosols. Generally, aerosols from biomass combustion processes can be divided into inorganic and organic aerosols. In the following the basic mechanisms leading to the formation of these two aerosol fractions are briefly summarised. Fig. 3 shows a simplified scheme of the most relevant processes involved.

Inorganic aerosols are formed from easily volatile species, released from the fuel to the gas phase during combustion. In this respect, K, S and Cl are the most relevant elements during the combustion of chemically untreated biomass fuels whereas Na and heavy metals (Zn and Pb) provide minor contributions. In the gas phase these species undergo reactions resulting in the formation of alkaline metal sulphates, chlorides and carbonates as well as heavy metal oxides. As soon as one of these compounds reaches a certain level of supersaturation (its concentration respectively vapour pressure exceeds the saturation vapour pressure), gas to particle conversion takes place. Supersaturation can be reached by the excessive formation of a specific compound (increase of its concentration) or by the
cooling of the flue gas (decrease of the saturation vapour pressure). Two gas to particle conversion processes, nucleation and condensation, are of relevance. If the concentration of submicron particles already existing in the flue gas is low, the formation of new particles in the size range of up to about 10 nm by nucleation takes place. As soon as existing particles can provide a sufficient amount of surfaces, condensation of ash forming vapours on these surfaces occurs. Then nucleation becomes of minor relevance and is finally almost totally suppressed. On their way with the flue gas through the furnace and the boiler section, the aerosol particles grow by coagulation processes. Finally, at boiler outlet the aerosol fraction usually shows an unimodal, almost normal logarithmic, particle size distribution. When applying chemically untreated wood fuels, the peak diameter of this particle size distribution is in the range of about 0.1 to 0.2 µm (a.e.d.). According to the formation mechanism described, the chemical composition of the fuel respectively the release behaviour of the easily volatile elements contained in the fuel, is the most relevant influencing factor regarding the formation of inorganic aerosols [8].

Organic aerosols represent the second fraction of fine particulate emissions. These particles are mainly due to incomplete soot combustion and to the condensation of unburned hydrocarbons during the cooling phase of the flue gas. Consequently, the formation of organic aerosols is a result of an incomplete flue gas burnout and can therefore be minimised by the optimisation of the combustion process. In fact it is well known, that in medium and large-scale combustion plants, where the flue gas burnout quality is usually very high (CO emissions <100 ppm and TOC emissions are almost not detectable), the formation of organic aerosols is nearly totally avoided [8].

3 METHODOLOGY

3.1 Overview

The methodological approach chosen is based on a considerable number of test runs performed with different types of small-scale biomass combustion systems and the subsequent evaluation and interpretation of the data achieved. Automatically fed pellet and wood chip furnaces and a manually fed log wood furnace have been investigated (only boilers, no stoves).

Concerning the automatically fed systems an important objective was to perform measurements at a test stand as well as field measurements in order to identify possible user-induced differences. Moreover, the test stand has been designed in a way that a flexible variation of the boiler load conditions could be achieved in order to simulate start-up, shut down and load change conditions.

During these test runs the concentrations and the particle size distributions of particulate emissions were measured downstream the boiler applying different particle measurement techniques. Furthermore, to define the operation mode of the furnace and the combustion conditions, the flue gas composition (O₂, CO, TOC, NO₃) was determined continuously and the most relevant operation parameters of the respective furnace were recorded. Selected particle samples taken during the test runs were forwarded to chemical analyses as well as electron microscopy to gain more detailed information about particle composition, shape and morphology.

3.2 Furnace types investigated

As already mentioned the investigations focused on different types of residential biomass heating systems. In this paper the results achieved for wood pellet and wood chip furnaces as well as a log wood furnace are presented.

Presently a huge variety of different residential biomass combustion systems is offered on the market. Especially concerning pellet boilers, which have shown a rapid development since their market introduction about 10 years ago [9], a considerable number of different fuel feeding strategies, furnace designs and process control options exists [10]. Therefore, it has been important to carefully select the systems investigated in order to identify possible differences between their technological features. 6 modern Austrian pellet furnaces have been tested. In order to apply the present state-of-the-art, only furnaces not older than 2 years have been selected. The nominal boiler capacity of all furnaces was in the range between 15 to 20 kW. All furnaces were equipped with automatic ignition systems, staged combustion, automated boiler cleaning systems as well as automated de-ashing systems. The furnaces differed concerning the following specific technological features:

- Feeding system: underfeed stokers, overfed burners, horizontally fed burners.
- Different grate systems.
- Furnace design: water cooled combustion chamber, combustion chamber insulated with refractory material.
- Control system: λ-control or furnace temperature control.
- One system has additionally been equipped with a flue gas condenser downstream the conventional boiler for latent and sensible heat recovery.

Furthermore, 3 state-of-the-art Austrian wood chip furnaces with nominal boiler capacities between 30 and 40 kW have been investigated. All furnaces were equipped with automatic ignition systems, staged combustion, λ-control, automated boiler cleaning systems as well as automated de-ashing systems. According to their technological features they differed as
follows:

- Feeding system: underfeed stoker or horizontally fed burner.
- Furnace design: water cooled combustion chamber or combustion chamber insulated with refractory material.

The log wood boiler tested had a nominal burner capacity of 30 kW and was equipped with a \( \lambda \)-control and an automated boiler cleaning system. In contrast to the automatically fed furnaces it had to be stocked manually with wood logs which were combusted via a modern downdraft system. In the lower part of the stock chamber the wood is gasified with primary air and the resulting flue gases are, by the addition of secondary air, burned in a subsequent cyclone burner chamber. The latter is insulated with refractory material. The ash has to be removed manually.

3.3 Testing campaigns and furnace operation

In total 6 testing campaigns at the test stand and 4 field testing campaigns have been performed with the different furnaces mentioned.

The furnaces tested at the test stand have been supplied and installed by the respective manufacturer. The furnace was taken into operation by the manufacturer and the test stand personnel was briefly instructed concerning the process control system. The test runs have been performed in absence of personnel from the manufacturer.

Concerning the field tests it is important to mention, that neither prior to nor during the testing campaigns optimisations or changes in the control system took place. The furnaces have been operated “as usual” at load conditions depending on the ambient temperature changes. No actions were undertaken to influence the heat demand of the central heating systems connected to the respective boiler. Consequently, the data gained from these test runs, are typical for the operation of the systems at the end users.

3.4 Setup of the test stand

A test stand was designed and installed at the Austrian Bioenergy Centre in Graz. The test stand provides the possibility to independently control the furnace load and therefore, full load and partial load operation as well as load changes, start-up and shut down procedures can be simulated. A measurement section downstream the boiler was designed to provide optimum conditions for the application of different particle sampling and measurement devices (see Fig. 4 and section 3.5). To overcome the pressure loss caused by this measurement section, an additional flue gas fan is implemented. By air addition at the end of the measurement section the pressure at boiler outlet is controlled and thereby usual stack conditions are simulated.

**Figure 4:** Measurement section of the test stand

3.5 Measurement and analyses methods applied

A significant number of different measurement and sampling technologies was applied in order to gain detailed information about the characteristics of PM emissions as well as about the operation conditions of the small-scale combustion systems investigated.

To define the performance of the respective combustion systems, characteristic process data such as furnace temperature, flue gas temperature, flue gas velocity and boiler load were recorded continuously. Moreover, the flue gas composition was measured and continuously recorded by using standard flue gas analyzers for \( \text{O}_2 \) (paramagnetic sensor), \( \text{CO} \) (NDIR), \( \text{NO}_x \) (CLD) and TOC (FID).

To characterise particulate emissions by their particle size distribution (PSD) and concentration in the flue gas, the following methods respectively equipment have been applied:

- Determination of the total fly ash concentration in the flue gas according to VDI 2066.
- Determination of the PSD and the concentration of aerosols in the flue gas with \( 9 \)-stage Berner-type low-pressure impactors (BLPI).
- Continuous determination of the PSD and the concentration of aerosols in the flue gas with an electrical impactor (ELPI 10 lpm).
- Determination of the TSP as well as PM\(_{10}\) and PM\(_{2.5}\) fractions with a PM\(_{10}\) impactor (Johnas II). At the test stand these measurement devices as well as the flue gas analysers have been connected to the measurement section. During field tests they were inserted in the flue gas duct downstream the boiler respectively into the stack.

In order to gain information about the chemical composition of the aerosol emissions selected BLPI samples were analysed applying the following methods:

- SEM/EDX (scanning electron microscopy / energy dispersive X-ray spectrometry)
- Wet chemical analyses concerning K, Na, S, Cl, Ca, Zn and Pb (pressurised multi-step digestion with HNO\(_3\)/HF/H\(_2\)BO\(_3\) and element detection with ICP-MS or AAS).

To get an indication about the \( \text{C} \)-content of the samples, also \( \text{C} \) was detected during the EDX-analyses. It has to be mentioned that results from C-measurements by EDX can only be seen as qualitative results. However, the results gained for K, Na, Zn, S and Cl from SEM/EDX and wet chemical analyses of the same impactor foils were compared with each other, and, for most of the elements, only deviations <20% occurred. Therefore, it was assumed that the C-concentrations gained from SEM/EDX are in an acceptable confidence interval. To distinguish between the organic C concentrations and the carbonate content of these particles, the carbonate content was calculated by taking into consideration, that K and Na are preferably bound as chlorides and sulphates and the remaining K and Na is bound as carbonates. A detailed description of the calculation model is given in [11].

As it has been mentioned in section 2.3, the chemical composition, i.e. the concentration of aerosol forming elements in the fuel, influences aerosol formation. Therefore, the fuels applied during the test runs have been analysed concerning the following parameters:
• ash content: method according to prCEN/TS 14775
• moisture content: determination of the weight loss at 105°C
• C, H, N-content: elemental analyser
• Cl: bomb combustion in oxygen; absorption in NaOH and detection by IC.
• Measurement: ion chromatography.
• S, Si, Ca, Mg, K, Na, Zn: pressurised multi-step digestion with HNO₃/HF/H₃BO₃ and element detection with ICP-MS or AAS

3.6 General remarks concerning the different particle measurement techniques applied
The results of each particle measurement technique applied provide a certain information depth. Total dust measurements for instance result in a concentration of the total fly ash emissions in the flue gas while with the PM₁₀ impactor information about 3 different size fractions (>10 µm, 2.5-10 µm and <2.5 µm) can be achieved. BLPI measurements provide particle concentrations for 9 particle fractions according to the cut diameters of the BLPI applied (0.0625, 0.125, 0.25, 0.5, 1, 2, 4, 8 µm aer.). All these techniques are based on gravimetric methods and are operated discontinuously. Therefore, their results represent mean values over the sampling time which is about 30' for the total dust sampling, about 20' for the PM₁₀ impactor and <5' for the BLPI. This fact has always to be considered when comparing the results gained with the different measurement techniques. Even if measurements with the 3 devices are started at the same time, fluctuations in the combustion process may lead to deviations of the results due to the different sampling durations.

The only quasi-continuous particle measurement device applied is the ELPI. It provides information about the number concentration and particle size distribution of particulate emissions in 11 size classes in the range between 0.034 and 9.97 µm (aer.) in 1-second intervals. Particle mass concentrations calculated from these number concentrations have to be treated with care, since some parameters which are not well known (shape and density of the particles in each size fraction) have to be considered. Therefore, in this paper, mass concentrations of particulate emissions are always based on the results gained from the discontinuous gravimetric methods applied. The ELPI results are mainly used to demonstrate and analyse changes of the aerosol emissions with respect to fluctuating operation and combustion conditions as well as to determine the number concentrations of aerosols in the flue gas.

3.7 Fuels used
The general description of the aerosol formation process in section 2.3 clearly indicates, that, as long as almost complete gas phase burnout conditions prevail, the composition of the fuel significantly influences the mass of aerosols formed. Therefore, the evaluation of results from wet chemical analyses of the fuels applied, as they are presented in Table I, is important for the interpretation of the results of the test runs.

In Austria wood pellets are standardised by the ÖNORM M 7135. Since only softwood pellets according to this standard have been applied, the deviations between the different samples analysed are in a small range. It can therefore be stated, that for all test runs with wood pellets comparable fuels have been used. This is especially true with respect to the aerosol forming elements.

Hardwood logs (beech) according to ÖNORM M 7132 (specifies terms and properties for the technological assessment of wood and bark as fuel) and ÖNORM CEN/TS 14961 (deciduous wood 1.1.2.1, P500, M20) have been applied for the test runs. P500 defines the size class (length: 500 mm +/-40 mm; diameter: 60–250 mm), M20 specifies the water content (<20% wt.% w.b.). As can be derived from Table I also for the wood logs used the deviations of the concentrations of aerosol forming elements are in an acceptable range.

Concerning the concentrations of ash forming elements, the wood chips used show a broad range of variations which is mainly due to the fact, that the fuel mixtures applied contained different amounts of hardwood, softwood and bark. Therefore, the fuel batches used during the different test runs with the three wood chip furnaces are listed and discussed separately.

Table I: Chemical characterisation of the fuels applied

<table>
<thead>
<tr>
<th></th>
<th>wood chips 1</th>
<th>wood chips 2</th>
<th>wood chips 3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>mean s</td>
<td>mean s</td>
<td>mean s</td>
</tr>
<tr>
<td>mc</td>
<td>27.6 0.4</td>
<td>17.4 0.8</td>
<td>23.9 0.6</td>
</tr>
<tr>
<td>ac</td>
<td>0.87 0.002</td>
<td>0.71 0.001</td>
<td>1.41 0.003</td>
</tr>
<tr>
<td>C</td>
<td>49.2 0.2</td>
<td>49.5 0.3</td>
<td>49.1 0.1</td>
</tr>
<tr>
<td>H</td>
<td>6.3 0.2</td>
<td>6.9 0.01</td>
<td>6.2 0.01</td>
</tr>
<tr>
<td>N</td>
<td>0.12 0.01</td>
<td>0.12 0.00</td>
<td>0.17 0.01</td>
</tr>
<tr>
<td>S</td>
<td>148.3 7.2</td>
<td>133.3 6.1</td>
<td>133.0 12.3</td>
</tr>
<tr>
<td>Cl</td>
<td>28.6 1.0</td>
<td>39.6 23.0</td>
<td>38.8 10.4</td>
</tr>
<tr>
<td>Ca</td>
<td>2,360 495</td>
<td>1,865 155</td>
<td>3,667 470</td>
</tr>
<tr>
<td>Si</td>
<td>432 123</td>
<td>320 59</td>
<td>931 482</td>
</tr>
<tr>
<td>Mg</td>
<td>299 12</td>
<td>216 20</td>
<td>309 53</td>
</tr>
<tr>
<td>K</td>
<td>972 24</td>
<td>557 41</td>
<td>1,187 40</td>
</tr>
<tr>
<td>Na</td>
<td>15.5 1.7</td>
<td>20.7 6.5</td>
<td>19.7 17.1</td>
</tr>
<tr>
<td>Zn</td>
<td>9.9 0.4</td>
<td>14.2 4.2</td>
<td>11.4 1.9</td>
</tr>
</tbody>
</table>

As can be seen from Table I the fuel moisture content increases from pellets over log wood to wood chips. The same is true for the ash content where even within the different wood chip batches analysed, remarkable differences occur.

Regarding the aerosol forming species mentioned in section 2.3, in all fuels K generally shows the highest
concentrations. The K-concentrations increase from softwood pellets over log wood to wood chips. Among the wood chips again an increase from batch 2 over batch 1 to batch 3 can be recognised. In all fuels tested S is the aerosol forming species with the second highest concentration followed by Cl. The S-concentrations also increase from softwood pellets over logwood to wood chips. The concentrations of other aerosol forming species such as Ca and Na are in all fuels by far smaller than the K-concentrations and therefore it can be expected, that K and consequently K-compounds (K$_2$SO$_4$, KCl and K$_2$CO$_3$) will play the dominating role during the formation of inorganic aerosols.

When adding up the concentrations of all relevant aerosol forming species in one fuel assortment (K, S, Cl, Na, Zn), the totals increase from wood pellets (558 mg/kg d.b.) over logwood (994.6 mg/kg d.b.) to wood chips (1,110 mg/kg d.b., mean value of all wood chip batches investigated). Consequently, wood chips show the highest and softwood pellets the lowest potential for inorganic aerosol formation.

With respect to the non-volatile ash forming elements Ca, Si and Mg the by far highest concentrations were determined for wood chips which can be explained by the larger bark fraction in this fuel.

4 RESULTS AND DISCUSSION

In this section the results from the measurement campaigns performed with the different small-scale biomass boilers are summarised. Firstly, data gained at stable load conditions are presented to provide a general overview over the particulate emissions of the different systems. Then, selected typical examples for particulate emissions at unstable load conditions are given to show how start-up procedures and load changes influence particle formation. Finally the particle size distributions and the chemical compositions of the PM emissions are presented and discussed. All gaseous and particulate emission data mentioned are related to dry flue gas and 13 vol.% O$_2$. Notations in mg/MJ are related to the net calorific value (NCV).

4.1 Total dust emissions at stable load conditions

In Fig. 5 the results of the total dust measurements performed at the different furnace types at full and partial load (±50% of full load) are summarised. The log wood furnace investigated shows with 8.8 mg/Nm$^3$ (5.7 mg/MJ) at full load and 6.8 mg/Nm$^3$ (4.4 mg/MJ) at partial load the lowest emissions. It has to be mentioned that these emission data for the log wood boiler are expected, that K and consequently K-compounds (K$_2$SO$_4$, KCl and K$_2$CO$_3$) will play the dominating role during the formation of inorganic aerosols. Moreover, no significant deviations related to stable combustion and load conditions during the test runs, are considerably lower which reflects the technological development achieved.

![Figure 5: Average total dust emissions of the furnaces investigated](image)

Explanations: Pe … pellet furnace; WC … wood chip furnace; LW … log wood furnace; field tests: Pe 3 and 4, WC 1 and 2; mean values and standard deviations of at least 3 single measurements

4.2 Fine particulate emissions at stable load conditions

In Fig. 6 the PM$_{10}$, PM$_{2.5}$ and PM$_{1}$ emissions determined with the BLPI during the same test runs are summarised. Comparing the data in Fig. 5 and Fig. 6 for pellet furnaces it can be derived, that the PM$_{10}$ emissions are close to the total dust emissions which means, that more than 90% of all particles are in the size range <10 µm (PM$_{10}$). Since Fig. 6 also shows that for pellet furnaces no significant differences between the PM$_{10}$-PM$_{2.5}$ and the PM$_{1}$ emissions occur, more than 90% of the total dust emissions are related to the PM$_{1}$ fraction.

About the same picture can also be found for wood chip furnace 1 and 2. Wood chip furnace 3 shows significantly higher total dust than PM$_{10}$ emissions resulting in a share of PM$_{10}$ on the total fly ash emissions of about 75%. Also the particle load in the size range between 1 and 10 µm is slightly higher for wood chip furnace 3.

For the log wood boiler, at full load, approximately 90% of the total dust emissions are related to PM$_{10}$ while during partial load 41% of the total dust emissions are related to PM$_{10}$.

Some details concerning these results of the PM emissions measurements are worth to be discussed in more detail. First of all, compared with medium and large-scale systems, where the total dust emissions...
usually consist of more than 50% coarse fly ash particles [8], the coarse fly ash emissions of the small-scale systems are extremely low. One reason therefore is that in small-scale combustion systems the fuel bed is less disturbed than in moving grate furnaces. This and the fact that the flue gas velocities in small-scale systems are by far lower than in larger scale systems causes a reduction of coarse fly ash entrainment from the fuel bed. A second reason is that the ash contents of pellets and log wood are comparably low. Only the fuel batch with the by far highest ash content (wood chip 3, see Table I) showed increased coarse fly ash emissions which underlines the influence of the ash content on coarse fly ash emissions.

A second interesting detail are the increasing PM10 emissions from the log wood furnace over the pellets furnaces to the wood chip furnaces. As mentioned in section 2.3, aerosol emissions can be divided into inorganic aerosols and organic aerosols. Since organic aerosols are a consequence of an incomplete gas phase burnout, elevated TOC emissions can be used as an indicator for the increased formation of these particles. During the full load test runs with all furnaces tested, the TOC emissions were almost not detectable (<3 ppm). Consequently, the difference concerning the fine particulate emissions between the different furnace types cannot be due to the formation of organic aerosols.

During the discussion of the fuel analyses data presented in Table I it has already been mentioned, that the wood chips used show higher concentrations of aerosol forming elements, especially K, than the pellets and the wood logs applied. In Fig. 7 the PM1 emissions measured at stable full load operation are plotted against the total concentration of aerosol forming species in the respective fuel. Since the emissions and the fuel data concerning pellets are within very narrow margins, mean values and standard deviations have been calculated from all measurement data gained from test runs with pellet furnaces. The diagram nicely documents how the PM1 emissions determined at pellet furnaces and wood chip furnaces increase with increasing concentrations of aerosol forming elements in the fuel. Consequently the composition of the fuel used and not the combustion technology applied is the main parameter responsible for the difference in the PM emissions of pellet and wood chip furnaces presented in Figure 5 and 6.

However, the results gained for the log wood furnace represent a significant outlier of this trend. The reason for this extreme deviation is assumed to be caused by a different release behaviour of aerosol forming species during log wood combustion. From former investigations it is known, that reducing atmosphere as well as low combustion temperatures can reduce the release of aerosol forming elements from the fuel and enhance the embedding of these elements into the bottom ash [12, 13]. It can be assumed, that during the main gasification/combustion phase reducing atmospheres and comparably low temperatures prevail in the wood logs and consequently, the release of aerosol forming species, especially of K, should significantly decrease during this phase. This phenomenon however, needs further and more detailed investigations.

Figure 6: PM10, PM2 and PM1 emissions determined for the different furnaces tested

Explanations: upper diagram: full load; lower diagram: partial load (=50% of full load); Pe … pellet furnace; WC … wood chip furnace; LW … log wood furnace; mean values and standard deviations have been calculated from at least 4 BLPI measurements

Figure 7: PM1 emissions vs. the concentrations of aerosol forming elements in the fuel applied

Explanations: fuel composition data: see Table I; PM1 emission data according to Fig. 5, full load; pellets: mean values including all BLPI measurements during full load operation at the pellet furnaces investigated; Pe … pellet furnaces; WC … wood chip furnace; LW … log wood furnace

Another interesting detail can be found when comparing the particulate emissions of the wood chip furnaces 1 and 2 at full load and at partial load operation, which indicate an increase of the emission for partial load. The operation data for partial load at these two furnaces show decreased furnace temperatures as well as increased TOC (up to 5 ppm) levels. Therefore, it can be assumed, that in this case the formation of organic aerosols due to incomplete combustion is responsible for the slightly increased PM emissions. This conjecture is also strengthened by the fact that calculations of the organic C content of the aerosol samples (see section 3.4 and 4.6) revealed higher values for partial load (up to 45 wt%) than for full load (up to 11 wt%).

4.3 Particulate emissions at unstable operation conditions

During load changes and especially during the start-up of a biomass furnace operation phases with
In the following typical examples for start-up phases and load changes for the pellet furnaces, wood chip furnaces and the log wood furnace tested are presented.

In Fig. 8 relevant operation data as well as gaseous and PM	extsubscript{1} emissions (gained from ELPI measurements) for a typical start-up phase of a pellet furnace are shown.

The horizontal lines in the PM	extsubscript{1} emission diagram represent the emission levels measured with the BLPI and the total dust measurement equipment whereas the length of the lines indicates the sampling period. Generally, the results gained from the discontinuous gravimetric measurement devices correspond well with the mean values of the ELPI measurement over the same period.

Especially at the beginning of the start-up phase the furnace temperatures are not high enough to provide sufficient burnout conditions. Therefore, increased CO and TOC emissions must be noted and consequently, due to the formation of organic aerosols, also the PM	extsubscript{1} emissions show a significant peak. As soon as the furnace temperatures are high enough for a complete combustion, the CO, TOC and PM	extsubscript{1} emissions simultaneously decrease. Subsequently, stable full load operation at low gaseous and particulate emissions prevails.

In the actual case it took about 30’ to reach full load (20 kW). During a 4-minute period elevated PM	extsubscript{1} emissions with a peak value of about 145 mg/Nm	extsuperscript{3} have been measured. The average emission for the whole 30’ start-up phase amounted to about 20 mg/Nm	extsuperscript{3} (13 mg/MJ), which is of course 65% higher than the PM	extsubscript{1} emissions for a typical start-up phase of a pellet furnace are shown.

The load change phases are followed by a longer period at full load. As mentioned in the previous section, wood chip furnace 3 showed slightly increased particulate emissions during partial load. As can be gathered from Fig. 9, the furnace temperature significantly decreased from full to partial load, in parallel the TOC emissions increase and consequently, also the PM	extsubscript{1} emissions increase due to the formation of organic aerosols.

Finally, during the shut down phase, again a short period with increased CO, TOC and PM	extsubscript{1} emissions which is mainly due to the decreasing furnace temperature, occurs.

In contrast to Fig. 8 the total dust emissions presented in Fig. 9 are higher than the PM	extsubscript{1} emissions. This is, as already mentioned, due to the higher amount of coarse fly ashes formed during this test run. The BLPI data however, are again in good compliance with the ELPI-data.

In Fig 9 operation and emission data of a typical load change from full to partial load during a test with wood chip furnace 3 are presented. With a rapid decrease of the boiler load at the beginning of the test (11.50) also the flue gas temperature decreased and a CO and TOC emission peak is recognised. In parallel with the TOC emission peak also a PM	extsubscript{1} emission peak occurs due to the formation of organic aerosols. Then (12.03) the boiler load is changed again to full load. During this phase no significant emission problems can be identified. The following second load change to partial load (12.20) also causes no significant TOC or PM	extsubscript{1} emission peak. Compared with the first load change also the furnace temperature drops to a less extent. This indicates that the process control system reacted on the second load change by far better than on the first one and therefore, this example underlines the importance of the process control system regarding particulate emission reduction.

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About the same start-up behaviour as described for pellet furnace 5 in Fig. 8 has been observed for all small-scale combustion technologies investigated.

A comparable load change behaviour has been observed for the pellet furnaces investigated. For pellet furnace 1, 2 and 5 load changes from full to partial load...
showed almost no effect on the PM emissions. Only for pellet furnace 6 an increase of the PM$_1$ emissions to an average value of 28 mg/Nm$^3$ (18 mg/MJ) had to be recognised which was mainly due to a non appropriate reaction of the process control system on the load change. Concerning load changes from partial load to full load, pellet furnace 1 and 6 showed slightly increased PM$_1$ emissions (28 respectively 29 mg/Nm$^3$, 18 respectively 19 mg/MJ). The differing behaviour of the different furnaces may possibly be attributed to the different technologies used as well as to the control systems applied.

Operation and emission data measured during the start-up phase as well as during a load change from full to partial load operation of the log wood boiler are presented in Fig. 10. Compared with a pellet or wood chip furnace, the start-up lasts significantly longer (about 60 minutes to reach 30 kW), however, the phase with elevated CO, TOC and PM$_1$ emissions at the beginning is still rather short (about 10 min.). As soon as optimum burnout conditions are reached, the PM$_1$ emissions remain at the low level already presented in Fig. 6 for stable load conditions.

![Figure 10: Emissions and operation data during start-up and load changes of the log wood boiler](image)

**Explanations:** measurements at test stand; FT … furnace temperature; P … boiler load; BLPI … Berner-type low-pressure impactor; ELPI … electric low pressure impactor; all concentrations related to dry flue gas; PM$_1$, CO, TOC emissions related to 13 vol% O$_2$.

At about 13.00 the load request is reduced to 50% of the nominal load. In contrast to automatically fed pellet or wood chip furnaces a manually fed log wood furnace has no possibility to adjust the fuel supply to changing load conditions. Therefore, the reaction of the control system on the load change causes a significant decrease of the furnace temperature which leads to a severe increase of the CO, TOC and PM$_1$ emissions. In the following the furnace temperatures periodically increase and decrease and also the emissions pulsate accordingly. This example nicely illustrates how rapid load changes can affect the performance of a log wood boiler and it also underlines the demand of a heat storage tank in order to smoothen changing load requests.

The example mentioned above represents an extreme case since for the log wood furnace load changes did usually not cause such strong disturbances, especially when the load change was carried out more carefully.

Either no or TOC emission peaks up to 50 mg/Nm$^3$ (32 mg/MJ) were observed for other load change phases investigated. The PM$_1$ emissions during these load changes were in the range between 5 and 9 mg/Nm$^3$ (3.2 and 5.8 mg/MJ), while during the extreme case shown in Fig. 10, the average PM$_1$ emissions increased to 12 mg/Nm$^3$ (8 mg/MJ) until stable combustion conditions could be reached again. This is however, also only a minor increase.

4.4 Average particulate emissions

The data presented in the previous sections clearly show, that start-up procedures generally cause increased PM emissions while the increase of emissions during load changes strongly depends on the performance of the control system as well as on the type of biomass heating system applied. However, from the data it can also be derived that modern small-scale biomass combustion units have the possibility to overcome instationary combustion conditions within shortest time. Therefore, the average PM$_1$ emissions for automated pellet furnaces and automated wood chip furnaces only slightly increase over a whole operation period including start-up and load changes. For the log wood boiler the evaluation of the ELPI data over a whole combustion period which lasted 7.5 hours (including start-up, full and partial load as well as load changes), showed an average particulate emission level of 8.4 (+/- 6.4) mg/Nm$^3$ (5.4 +/- 4.1 mg/MJ).

4.5 Particle size distributions of PM$_1$ emissions

In Fig. 11 the PSDs of the PM$_1$ fractions determined with the BLPI (mass size distributions) and the ELPI (number size distributions) at stable full load conditions are shown. The shapes of the mass PSDs for all furnaces tested are comparable and show a peak in the size range of about 0.1 - 0.2 µm. With increasing concentration of aerosols in the flue gas usually their particle size slightly increases due to enhanced coagulation effects and surface condensation [8]. Consequently, the aerosols emitted from the log wood boiler show the smallest distribution peak and the smallest particle size.

Furthermore, it can be derived from Fig. 11 that for the pellet furnaces and the log wood furnace close to a particle size of 1 µm the PSD approaches 0 which is in agreement with the results presented in Fig. 6 indicating that almost the whole particulate emissions measured are related to PM$_1$. Only for the wood chip furnace a very small amount of particles can be found in the size range above 1 µm.

Despite of this no significant differences between the combustion technologies tested can be seen. However, the evaluation of the PSDs determined during operation phases with elevated particulate emissions (start-up and load changes) show a slight shift to larger particle sizes (by up to about 0.1 µm) which again is assumed to be due to an increased efficiency of agglomeration processes due to the higher number of particles formed and due to increased surface condensation of organic aerosols during the cooling phase of the flue gas.
4.6 Chemical composition of the aerosol emissions

Samples taken with the BLPI were forwarded to SEM/EDX and wet chemical analyses. In Fig. 12 the composition of the inorganic part of the aerosol emissions gained from the results of the wet chemical analyses of the BLPI-samples are presented.

No significant difference between particles sampled during full load and partial load have to be mentioned.

As already expected from the evaluation of the fuel analyses, K is the dominating element in the aerosol fraction followed by S and Cl. Consequently, K2SO4 and KCl are the most relevant constituents of the aerosols. From the comparison of the K, S and Cl concentrations it can be derived, that the amount of S and Cl is not high enough to bind all the K present. The remaining K is therefore bound as K2CO3. Besides K, S and Cl, minor amounts of Na and Zn can be found in all samples analysed.

The aerosol composition also nicely resembles the fuel composition concerning the S/Cl ratio. For wood chips and log wood the Cl-concentrations in the fuel are significantly lower than the S-concentrations while for the pellets these two elements show about the same concentration level. Consequently, also the aerosols formed during pellet combustion show significantly higher Cl but lower S concentrations than the aerosols from the other fuels.

These inorganic aerosols form the basic level of aerosol emissions and therefore represent the minimum emission that can be achieved at a given release of aerosol forming species from the fuel at optimum burnout conditions.

The comparison of the results gained from the wet chemical analyses with the results from the SEM/EDX-analyses (including the C-determination) indicates, that at stable operation conditions, for the pellet furnaces, the concentration of non-carbonate carbon is in a range <11 wt% for full and <34 wt% d.b. for partial load. The consecutive values for the wood chip furnaces are <11 and < 45 wt% d.b. and for the log wood boiler <4 and <10 wt% (see Fig. 13). During instationary combustion conditions this share increases and reaches, for instance during load changes at pellet furnaces levels of up to 60 wt.% d.b. For start-up procedures (e.g. PM emission peak of 145 mg/Nm³ (94 mg/MJ) in Fig. 8) or during phases with poor burnout conditions (e.g.: PM emission peak of 75 mg/Nm³ (48 mg/MJ) in Fig. 10) organic carbon and soot can make up to 90% of the composition of the aerosol emissions.
5 CONCLUSION AND RECOMMENDATIONS

Test stand and field measurement campaigns with 6 automatically fed pellet furnaces, 3 automatically fed wood chip furnaces and one log wood boiler have been performed in order to characterise particulate emissions by their concentration in the flue gas, their particle size distribution and their chemical composition. Only state-of-the-art combustion units have been applied. The test runs have been performed at stable full and partial load conditions but also during unstable conditions as start-up and load changes.

Concerning gaseous emissions (CO, TOC) it has already been known, that the R&D efforts undertaken during the last years, resulted in a significant decrease of emissions. Concerning particulate emissions, especially during the last years, resulted in a significant decrease of particulate emissions during stable full and partial load conditions but also during unstable conditions as start-up and load changes.

Concerning particulate emissions, especially during load changes phases with incomplete gas phase burnout can occur. Again, the formation of organic aerosols and therefore, an increase of the particulate emissions is the consequence. For automatically fed systems (pellet and wood chip furnace) it was found, that the quality of the process control system applied strongly influences particle formation during this period. If the process control reacts fast enough on changing load requests, almost no emission peaks have to be recognised. In the opposite case, PM emissions of up to 200 mg/Nm³ (133 mg/MJ) a significant decrease of PM emissions can be stated for state-of-the-art combustion systems. However, there is still some potential for optimisation left.

Another important primary measure for the reduction of particulate emissions is the combination with heat storage tanks in systems which show PM emissions in a range up to 200 mg/Nm³ (133 mg/MJ) a significant decrease of PM emissions can be stated for state-of-the-art combustion systems. However, there is still some potential for optimisation left.

Generally, optimised furnace geometries and appropriate air staging concepts provide the basis for a combustion at the possibly lowest CO and TOC emissions which is a prerequisite for achieving low particulate emissions. For automatically fed systems, the further optimisation of the process control system applied can lead to a further decrease of particulate emissions. If the process control system is able to avoid operation phases with insufficient burnout conditions, especially during load changes and keeps these phases during start-up as short as possible, the formation of organic aerosols can be minimised.

Regarding log wood boilers, the combination with heat storage tanks is an appropriate measure. With heat storage tanks rapid load changes can be avoided, and therefore, unstable operation conditions, as they will happen in any case during a load change from full to partial load, can be avoided.

Another important primary measure for the reduction
of particulate emissions from small-scale biomass furnaces is related to the fuel chemistry. With decreasing concentrations of aerosol forming species in the fuel also the inorganic aerosol emissions decrease. Therefore, in small-scale biomass combustion systems, for which no appropriate secondary measures (filters) for aerosol precipitation are available on the market, only fuels with comparably low concentrations of these elements should be used. Consequently the utilisation of wood fuels, preferably softwood, should be favoured instead of the utilisation of herbaceous or agricultural fuels.

When exhausting all potentials of primary measures for the minimisation of fine particulate emissions from small-scale biomass combustion (optimised combustion to avoid the formation of organic aerosols and utilisation of softwood), emission levels of about 15 mg/Nm³ (10 mg/MJ) for pellet and wood chip furnaces and even lower values for log wood furnaces could be reached. This represents, compared with old combustion systems of course a significant improvement, however, compared with natural gas and oil fired combustion units, these emissions are still one order of magnitude higher. The results of the measurements performed clearly indicate, that PM emissions from optimised biomass combustion plants are dominated by K-salts. First results of research projects indicate that the toxicity of such particles is lower than for diesel soot. Therefore it is of great importance to further investigate the health risks caused by inorganic aerosol emissions from biomass combustion in order to find out, if additional, secondary measures are needed to further decrease the emissions, or if the toxicity of K-salts is at a level low enough, that the inorganic particulate emissions from an optimised small-scale biomass combustion plant are acceptable.

Initiatives to start appropriate projects on this topic are presently on-going at the Austrian Bioenergy Centre, Graz, as well as at the Institute for Resource Efficient and Sustainable Systems, Graz University of Technology.

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7 REFERENCES