

EVALUATION OF PARAMETERS DETERMINING PM EMISSIONS AND THEIR CHEMICAL COMPOSITION IN MODERN RESIDENTIAL BIOMASS HEATING APPLIANCES

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ABSTRACT: In this paper the results of a project performed at the Austrian Bioenergy Centre and the Institute for Process Engineering, Graz University of Technology, are presented, which focused on the investigation of particulate emissions from modern residential biomass heating systems. Pellet, wood chip and logwood boilers as well as a tiled stove have been tested during test stand and field measurement campaigns. The results of the project show, that modern residential biomass combustion appliances show significantly reduced particulate emissions compared to old systems. Based on the results PM₁₀ emission factors of 20 mg/MJ for pellet and logwood boilers, 20-30 mg/MJ for wood chip boilers and 40-55 mg/MJ for tiled stoves can be proposed. Moreover, parameters influencing PM emissions such as the chemical composition of the fuel, the burnout quality and the furnace temperature have been identified and their effect on the mass and chemical composition of PM emissions has been evaluated.

Keywords: emissions, aerosols, fly ash, combustion

1 INTRODUCTION

Biomass combustion presently plays an important role concerning residential heating in many European countries. As a recent survey [1] has revealed between 10 and 30% of the residential heating demand in Austria, Germany, Sweden and Finland is provided by biomass combustion systems. Presently mainly old systems are applied.

On the other side, during recent years residential biomass combustion systems have been more and more criticised due to their comparably high particulate matter (PM) emissions. From emission inventories it is well known that old residential biomass heating systems significantly contribute to PM₁₀ emissions. In this context organic carbon and soot contained in these PM emissions are assumed to cause adverse health effects.

However, it is also known, that R&D work performed during the last years, has significantly increased the technological standard of small-scale biomass heating plants and therefore also reduced their particulate emissions. In order to quantify and characterise PM emissions from state-of-the-art small-scale biomass combustion appliances, a project has been performed at the Austrian Bioenergy Centre, Graz, Austria, and the Institute for Process Engineering, Graz University of Technology, during the last 3 years. The project was based on a cooperation of these two research institutions with 9 Austrian biomass furnace and boiler manufacturers. It focused on the determination of PM emissions from state-of-the-art residential biomass heating systems, at their physical and chemical characterisation as well as on the investigation of relevant parameters influencing the formation and the chemical composition of these PM emissions. The results of this project are presented in this paper.

2 FUNDAMENTALS OF PARTICULATE EMISSION FORMATION IN BIOMASS COMBUSTION

Particulate matter (PM) emissions in biomass combustion generally have to be divided into coarse

particles (>1 µm) and fine particulate emissions, the so-called aerosols (particles <1 µm or PM₁). Both fractions together form the emission of total suspended particulate matter (TSP).

Coarse fly ash emissions are due to the entrainment of fuel and ash particles from the fuel bed with the flue gas. In medium and large-scale applications coarse fly ashes provide the major contribution to the total PM emissions while in residential biomass heating systems their share on the PM emissions is low [2].

Concerning the aerosol fraction, it has to be distinguished between inorganic aerosols as well as organic aerosols and soot particles. Inorganic aerosols mainly result from the release of easily volatile ash forming species from the fuel to the gas phase. During the combustion of chemically untreated biomass mainly K, S and Cl as well as, to a minor extent, Na and easily volatile heavy metals such as Zn and Pb are involved in this process. Subsequently they undergo chemical reactions and their compounds form submicron particles by gas to particle conversion mainly during the cooling of the flue gas [2]. Moreover, the entrainment of small amounts of submicron refractory oxide particles (CaO, SiO₂ and MgO) from the fuel bed contributes to inorganic aerosol formation.

Carbonaceous aerosols are a result of incomplete burnout and originate from the condensation of organic vapours (organic carbon emissions) as well as of incompletely oxidised soot particles (elemental carbon emissions). With decreasing burnout quality of the flue gases their contribution to the PM emissions significantly increases.

3 APPROACH

Testing campaigns with different modern (manufactured not before 2004) Austrian small-scale biomass combustion systems have been performed. 6 pellet boilers and 4 wood chip boilers, all equipped with staged combustion, automatic feeding, ignition and de-ashing systems as well as automatic combustion control systems have been selected. Moreover, 2 manually fed

logwood boilers equipped with staged combustion and automatic control systems and 1 tiled stove, designed according to the current guidelines of the Austrian Tile Stove Association have been tested. The pellet, wood chip and logwood boilers have thereby been chosen in a way that all market relevant differences concerning technological features (combustion technology, cooled and isolated furnaces, different control strategies) could be considered. The nominal thermal capacity of the systems was in the range between 12 and 35 kW.

Test runs have been performed at a specially designed test stand at the laboratory facilities of the Austrian Bioenergy Centre (ABC) in Graz (with all types of combustion systems) as well as during field tests (with pellet and wood chip boilers). The test stand has been planned and equipped to be able to simulate different load conditions (full load, partial load, load changes, start-up and shut down procedures) and therefore operation at stationary as well as transient load conditions could be investigated.

The combustion systems have been provided by the manufacturers engaged in the project. They were installed at the test stand and started up by the respective manufacturer. A short (1-day) initial training of the test stand personnel followed. During the subsequent testing phase no changes or optimisations of the control systems have been performed.

During the field measurements the combustion systems investigated have been operated at the load conditions determined by the ambient temperature. Depending on the climatic constraints full and partial load operation phases could be tested. The plants have been tested at usual operation conditions which means, that no changes or optimisations of the control systems have been performed.

Softwood pellets according to ÖNORM M 7135 as well as wood chips and logwood according to ÖNORM M 7132 were used as fuels.

The test programme was based on the following sampling, measurements and analyses procedures:

- Fuel characterisation: fuel analyses concerning moisture content, ash content as well as C, H, N, Ca, Si, Mg, K, Na, S, Cl and Zn.
- Evaluation of operation data such as load, furnace temperature and flue gas temperature in order to define the operation mode and the performance of the respective combustion plant.
- Flue gas analyses (O_2 , CO_2 , CO, OGC, NO_x) and PM emission measurements were performed downstream boiler/stove outlet.
- Concerning PM emissions TSP was measured and the concentration and particle size distribution of aerosols was determined with 9-stage Berner-type low-pressure impactors (BLPI).
- On-line determination of the particle size distribution and number concentration of aerosols in the flue gas was performed with an electrical impactor (Dekati ELPI 10 lpm).
- Selected PM samples taken with the BLPI were forwarded to SEM/EDX analyses as well as chemical analyses concerning the parameters total inorganic carbon (TIC), non-carbonate-carbon (sum of organic and elemental carbon) as well as Ca, Si, Mg, K, Na, S, Cl and Zn.

3 RESULTS ACHIEVED

3.1 TSP and PM_{10} emissions at stable load conditions

Firstly, a brief overview over the PM emissions measured during the test runs is given. Thereby, the automatically controlled boilers and the stove tested are discussed separately.

It has to be pointed out, that for the pellet and wood chip boilers tested no significant differences concerning the results gained from the test stand and the field measurement campaigns occurred. This is mainly due to the highly developed control systems which widely avoid operating errors.

In Fig. 1 average TSP and PM_{10} emissions determined during stable full load operation are presented. In the diagram mean values and standard deviations from test stand as well as field measurement campaigns are summarised. Since the pellet boilers tested at the test stand as well as during the field measurements showed no significant deviations concerning their PM emissions, these results are summarised in one box plot [3].

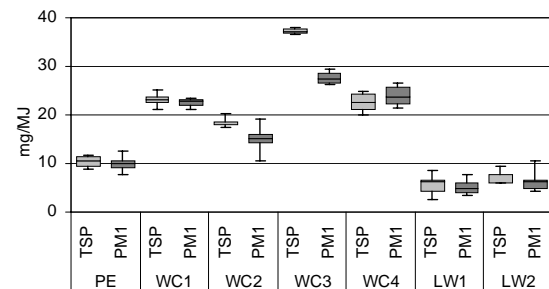


Figure 1: TSP and PM_{10} emissions of different residential biomass heating systems at full load operation. Explanations: Pe ... pellet boiler; WC ... wood chip boiler; LW ... logwood boiler

From Fig. 1 can be revealed that the TSP emissions from the modern pellet boilers tested are in the range below 13 mg/MJ (20 mg/Nm^3 , dry flue gas, 13 vol% O_2) while wood chip boilers show emissions of up to 30 mg/MJ (50 mg/Nm^3) and logwood boilers of in average 8 mg/MJ (10 mg/Nm^3). These TSP emissions are significantly lower than the emissions reported for older systems which amount up to 130 mg/MJ (200 mg/Nm^3) [4].

Fig.1 also shows that for pellet and logwood boilers as well as for two of the wood chip boilers tested the PM_{10} emissions almost equal the TSP emissions. More than 90% of the TSP emissions are related to the PM_{10} fraction. Two wood chip boilers showed significant coarse fly ash emissions amounting to about 25% of the TSP emitted. This can be explained with the comparably high ash contents of the fuels applied during these test runs (1.4 respectively 1.6 wt% d.b.). Moreover, the results of the measurements have revealed, that almost no particles in the size fraction between 1 and $10 \mu\text{m}$ are emitted, which means that PM_{10} almost equals PM_{1} .

As already mentioned, no significant differences concerning the PM emissions of the different pellet boilers could be detected even though applications with different furnace concepts (cooled and isolated furnace), combustion concepts (underfed stoker, overfed burner, horizontally fed burner) and control concepts (lambda

control, temperature control) have been tested. The same observations could be made for the two different logwood boilers. For the wood chip boilers however, significant deviations concerning the PM emissions occurred, which turned out not to be due to the combustion technologies applied but to differences in the compositions of the fuels used (see section 3.2).

Natural draught combustion systems such as the tiled stove generally show higher gaseous and particulate emissions due to a lack of any advanced combustion control mechanism. The mean PM₁ emissions over the whole batch amounted to 52 mg/MJ (80 mg/Nm³) which is significantly lower than values reported for older systems. Also at the tiled stove more than 90% of the TSP emissions were related to the PM₁ fraction.

3.2 Parameters influencing PM₁ emissions as well as their chemical composition

The evaluation of the results revealed three main parameters influencing PM₁ emissions:

- the concentration of inorganic aerosol forming species (especially K) in the fuel,
- the burnout quality,
- the furnace temperature.

In Fig. 2 the relation between the total concentration of inorganic aerosol forming species in the fuel (total of K, Na, S, Cl and Zn) and the PM₁ emissions measured at stable full load operation is shown. In pellet, wood chip and logwood boilers almost complete burnout characterised by low CO (<300 ppm) and OGC (<5 ppm) emissions prevailed during these operation phases. Consequently, mainly inorganic aerosol formation took place. Concerning the tiled stove only data from the main combustion phase, which are best comparable with stable conditions in automatically controlled combustion systems are considered. However, as it will be shown later, these particles also contain about 75% of organic species and soot.

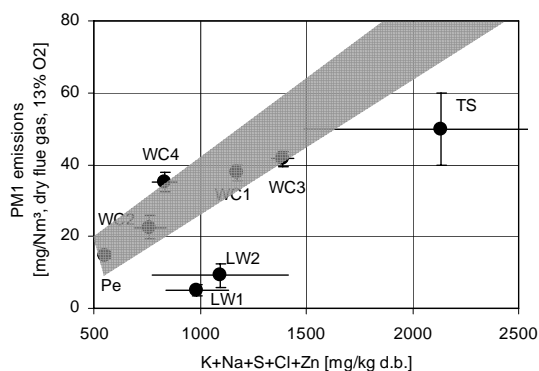


Figure 2: PM₁ emissions at full load operation vs. the concentration of aerosol forming elements in the fuel
 Explanations: PM₁ emissions related to dry flue gas and 13 vol.% O₂; Pe ... pellet boilers (average over all test runs performed); WC ... different test runs with wood chip boilers; LW ... logwood boilers; TS ... tiled stove; the data represent mean values and standard deviations; d.b. ... dry basis

As it can be seen, a trend of increasing PM₁ emissions with increasing contents of easily volatile ash forming elements in the fuel prevails for pellet and wood

chip boilers. This trend also explains the varying PM₁ emissions determined for the wood chip boilers tested and confirms that they are not due to different combustion technologies but to varying fuel quality.

The data for logwood boilers and the tiled stove however represent an exception to this trend. The K-release from the fuel to the gas phase seems to be hindered under the constraints of logwood combustion in boilers and stoves resulting in lower inorganic PM₁ emissions. 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 [5, 6]. 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.

A second relevant parameter influencing aerosol formation is the burnout quality of the flue gas. It affects both, the PM₁ emissions as well as their chemical compositions. In Fig. 3 the non-carbonate carbon (NCC - total of organic carbon and soot) concentrations of the PM₁ emissions are plotted against the OGC emissions. The data are taken from a test runs with a wood chip boiler at changing load conditions. From the data it can easily be derived that with increasing OGC emissions the NCC content of the aerosol particles formed significantly increases. This is due to the fact, that hydrocarbons, which are released from the fuel and which are not completely destroyed during gas phase burnout, condense during the cooling of the flue gas in the boiler respectively stove and form submicron organic particles or condense on existing particle surfaces. Moreover, incompletely oxidised soot particles contribute to these NCC emissions. Elevated CO and OGC emissions (>1,000 ppm respectively >10 ppm) are a good indicator for increased formation of NCC emissions. This aerosol formation mechanism is mainly observed during transient operation phases (start-up at pellet, wood chip and logwood boilers as well as the ignition and main combustion phase of stoves).

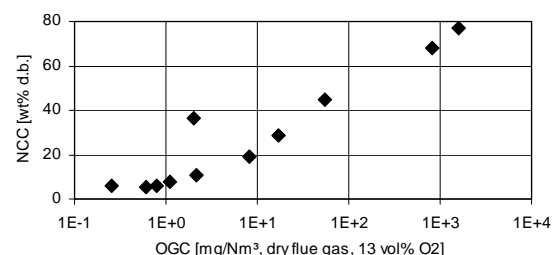


Figure 3: Relation between gaseous OGC emissions and the NCC content of aerosol emissions
 Explanations: NCC ... non carbonate carbon (organic carbon and soot); d.b. ... dry basis; data from a test run with a wood chip boiler

Moreover, the furnace temperature in the burnout zone of a furnace/stove can influence the NCC emissions. In Fig. 4 the PM₁ emissions as well as the NCC concentrations of aerosols are plotted against the furnace temperature in the burnout zone (after injection

of the secondary combustion air).

From these data it can be revealed that the PM₁ emissions increase with decreasing furnace temperature due to increased NCC emissions (this statement is valid for furnace temperatures <750°C). Besides the fact that usually with decreasing furnace temperature the burnout quality also decreases, increased soot emissions are also expected to be mainly responsible for this result. These findings are of special relevance regarding the partial load operation of boilers where lower combustion temperatures than at full load usually occur. It has been observed, that during these phases the burnout quality is comparable with full load operation (low CO and OGC emissions) but the PM emissions are elevated. This leads to the conclusion, that high CO and OGC emissions are in any case a good indicator for increased formation of organic aerosols and soot while low CO and OGC emissions do not necessarily exclude the formation of NCC particles, especially at low furnace temperatures and if furnace cooling is applied.

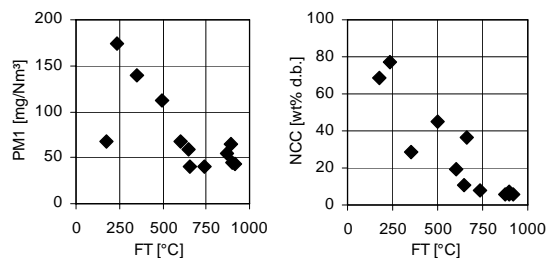


Figure 4: PM₁ emissions and their NCC content vs. the furnace temperatures during wood chip combustion
 Explanations: NCC ... non carbonate carbon (organic carbon and soot); PM₁ emissions related to dry flue gas and 13 vol% O₂; d.b dry basis; FT ... furnace temperature

3.3 Aerosol formation in different types of residential biomass combustion systems

The influencing parameters presented in the previous section are valid for all types of biomass combustion devices. However, their effects on PM₁ formation significantly vary depending on the type of combustion device investigated. It must generally be distinguished between 3 different cases:

- automatically fed and controlled systems (pellet and wood chip boilers),
- manually fed automatically controlled systems (log wood boilers),
- manually fed not controlled systems (stoves, tiled stoves).

In Fig. 5 typical operation and emission data recorded during the start-up phase of a pellet boiler are presented. At the beginning, when the furnace temperatures start to increase, the combustion conditions are not good enough (high excess oxygen level, low furnace temperatures) to achieve a complete burnout. Consequently, during this phase CO, OGC and PM₁ emission peaks occur. As described above, the PM₁ emissions during this phase are mainly due to the formation of organic aerosols and soot. As soon as the furnace temperature has reached a sufficiently high level for an almost complete burnout, which usually happens after some minutes, the CO, OGC and PM₁ emissions

rapidly decrease and reach the same level as determined at stationary full load conditions. For the remaining period until full load is reached, no further emission peaks are usually recognised. Even if the emission peaks at the beginning of the start-up phase amount up to the 10 fold value of the emission at stationary operation, the average emissions related to the whole start-up phase until full load is reached (up to 30 min. for the pellet boilers and up to 40 min. for the wood chip boilers tested) amounted up to 200% (pellet boilers) and up to 630% (wood chip boilers) of the emissions at stationary conditions.

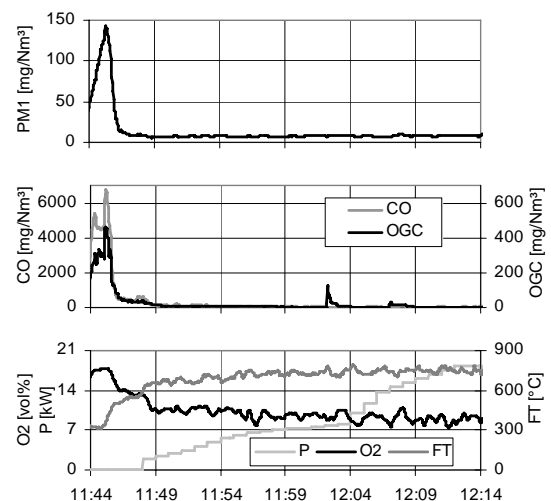


Figure 5: Aerosol, CO and TOC emissions during start-up of a pellet furnace

Explanations: particulate, CO and OGC emissions related to dry flue gas and 13 vol% O₂; O₂ concentrations related to dry flue gas; P ... load; FT ... furnace temperature

During subsequent phases with stable full or partial load operation conditions almost constant emissions in the ranges as presented in Fig. 1 were detected. During load change phases sometimes short CO, OGC and PM₁ emission peaks were recognised. These emission peaks are mainly due to short disturbances of the combustion process due to the changing load conditions, and their occurrence strongly depends on how fast the process control system of the respective combustion plant reacts on the load change.

The same start-up behaviour could be observed for manually fed automatically controlled systems (logwood boilers). However, due to the manual ignition and the larger size of the wood logs compared to wood chips and pellets, the emission peak at the beginning of the start-up phase was more pronounced. The PM emissions measured during continuous full and partial load operation conditions were lower than at the pellet and wood chip boilers, which is due to the reduced release of inorganic aerosol forming compounds during this phase (see section 3.2). A significant difference could be detected concerning the load change behaviour of manually fed systems. While increasing loads caused no emission problems load changes from full to partial load led to significant emission peaks. This is due to the reason, that batch burning devices cannot reduce the

amount of fuel in the fuel bed as automatically fed burners can do. Therefore, these systems cannot react appropriately on decreasing load demands. For that reason, logwood boilers should only be applied in combination with heat storage tanks in order to avoid rapid load changes from full to partial load. Another difference compared to the manually fed systems was observed during shut down (burnout phase). Here, a phase of usually more than 30 min. occurs during which the furnace temperatures decrease and the CO emissions increase. However, since only charcoal burnout prevails during this phase, no significant increases of OGC and NCC-particle emissions were observed.

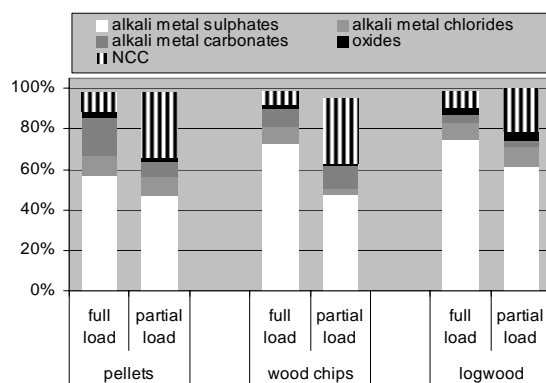


Figure 6: Average chemical composition of PM₁ emissions at full and partial load

Explanations: NCC ... non carbonate carbon (total of organic carbon and soot); data in wt% d.b.; partial load: 50% of full load

For automatically fed and manually fed automatically controlled systems, the PM emissions are mainly determined by the emissions at stable load operation since the emission peaks during start-up and load changes are comparably small. In Fig. 6 the chemical composition of particles emitted from pellet, wood chips and logwood boilers at stationary full and partial load (50% of full load) operation are presented. From the diagram can be derived that the aerosol emissions at full load are dominated by inorganic salts (more than 90 wt% of the total emission) while NCC compounds only amount to less than 10%. During partial load operation however, the composition shifts to a NCC share of up to 40 wt%. This shift is mainly due to the reason, that during partial load operation the furnace temperatures significantly decrease since most systems operate with cooled furnace walls. Therefore, the release of inorganic ash forming compounds also decreases but the particulate NCC emissions increase due to the lower temperatures and worse burnout conditions. Since the total PM emissions in most cases remain at about the same level, these two effects compensate each other regarding the total mass of particles formed.

PM₁ emission formation in manually fed not controlled batch burning devices as a tiled stove significantly differs from automated systems. One combustion batch is characterised by three different phases, the ignition, the main combustion and the burnout phase. The ignition phase turned out to be the most problematic in terms of PM₁ emissions, since low

furnace temperatures and high O₂-levels lead to incomplete combustion and thus the formation of organic aerosols and soot. During this phase average PM₁ emissions of up to 130 mg/MJ (200 mg/Nm³) have been determined while during the subsequent main combustion phase the emissions decreased significantly and remained at a comparably low level during the burnout phase. However, due to the comparably low furnace temperatures in these systems, the higher excess oxygen levels and the fact, that no combustion control mechanisms exist, the CO, OGC and consequently also the PM emissions measured during the main combustion phase showed with on average 26 mg/MJ (40 mg/Nm³) significantly higher values than at automatically controlled combustion systems. Consequently also the particle composition significantly differs from the one of automatically controlled systems. In Fig. 7 the particle compositions determined during the ignition and the main combustion phase of the tiled stove investigated are presented. It can be seen that in both cases NCC emissions dominate. While during the ignition phase more than 90% of the particles consist of NCC compounds their amount decreases during the main combustion phase to approximately 75%.

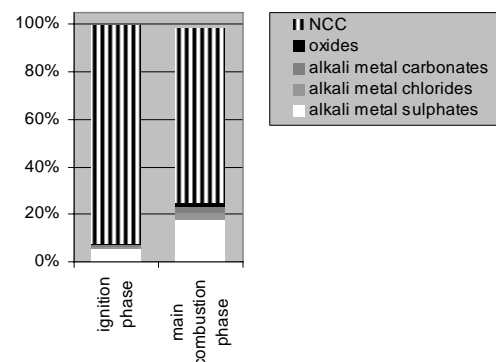


Figure 7: Average chemical composition of PM₁ emissions of a tiled stove

Explanations: NCC ... non carbonate carbon (total of organic carbon and soot); data in wt% d.b.

4 CONCLUSIONS AND RECOMMENDATIONS

Comprehensive measurement campaigns with modern automatic pellet, wood chips and logwood boilers as well as a tiled stove have been performed in order to investigate and characterise their PM emissions. The results of the measurements clearly show that state-of-the-art residential biomass heating systems operate at significantly lower PM emissions than older systems. The PM emissions are clearly dominated by the PM₁ fraction which amounts in almost all cases to more than 90% of the TSP emission. Since almost no particles were found in the size class between 1 and 10 μm, PM₁ almost equals PM₁₀. Considering the whole operation cycle including transient and continuous operation phases, the following PM₁₀ emission factors can be proposed: pellet boilers operated with softwood pellets and logwood boilers: 20 mg/MJ; wood chip boilers: 20-30 mg/MJ (depending on the K-content of the fuel); tiled stoves: 40-55 mg/MJ. These emission factors are significantly lower

BRUNNER Thomas, BÄRNTHALER Georg, OBERNBERGER Ingwald, 2008: Evaluation of parameters determining PM emissions and their chemical composition in modern residential biomass heating appliances. In: Proc. of the int. Conf. World BIOENERGY 2008, May 2008, Jönköping, Sweden, ISBN 978-91-977624-0-3, pp.81-86, Swedish Bioenergy Association (Ed.), Stockholm, Sweden

than the ones determined for older systems in previous studies (up to 130 mg/MJ).

However, the work performed has also shown that further potentials for PM emission reductions exist. During furnace design of automated systems it should be taken care, that an optimised mixing of the flue gases with the secondary combustion air can be secured during full and partial load operation in order to improve the burnout. Moreover, the design of the furnaces should be optimised in a way that also at partial load operation sufficiently high furnace temperatures can be maintained in order to avoid increased NCC emissions. A further optimisation of the control system, which especially focuses on the quick implementation of appropriate measures for burnout improvement during start-up and load changes can also help to reduce PM emissions.

Regarding stoves a basic optimisation guideline is to implement measures which support high combustion temperatures and a good mixing of the flue gas with the combustion air. However, especially at manually fed batch combustion systems the user behaviour significantly influences emission formation. Therefore, appropriate user training with respect to correct filling of stoves and fuel ignition is the first and most important step towards lower PM emissions.

When comparing the emission factors proposed for state-of-the-art systems with emission factors published for old systems, the high potential for emission reduction by replacing old by new systems becomes obvious. In Austria for instance more than 75% of the PM₁₀ emissions from domestic wood combustion could be omitted by this measure. In addition, a changeover from old to new systems would not only reduce the PM emissions but also their chemical composition. The emissions of organic aerosols and soot particles which are assumed to be of higher relevance concerning health effects than inorganic salts could be significantly reduced by this measure.

Since the technical optimisation potentials of modern automatic residential biomass combustion systems are almost exhausted, a further significant reduction of PM emissions must be based on the application of filters or the development of new combustion systems. Concerning particle precipitation devices for small-scale biomass combustion systems high R&D efforts are still needed, especially in order to increase their availability and to decrease their costs.

5 ACKNOWLEDGEMENTS

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