REAL-LIFE EMISSION FACTOR ASSESSMENT FOR BIOMASS HEATING APPLIANCES AT A FIELD MEASUREMENT CAMPAIGN IN STYRIA, AUSTRIA

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ABSTRACT

Biomass combustion is a major contributor to ambient air pollution. Thus, knowing the real-life emissions of biomass heating systems is crucial. Within the project Clean Air by biomass a field measurement campaign was conducted. 15 biomass heating appliances were tested in households at the end user according to their usual operation. Emission factors for gaseous and particulate emissions, as well as for the genotoxic and carcinogenic substance benzo(a)pyrene, were evaluated and compared to current proposed European and Austrian emission factors used for emission inventories. Moreover, the shares of particles and benzo(a)pyrene in hot and cooled flue gas were determined. Results showed a high variability of emissions in the field. Highest values and ranges occurred for room heaters (TSPtotal: 226 mg/MJ). Biomass boilers showed clearly lower emission factors (TSPtotal: 184 mg/MJ) in the field than room heaters and also than the proposed European and Austrian emission factors, in many cases. Emission factors for tiled stoves showed a similar trend (TSPtotal: 67 mg/MJ). The share of condensable particles in the flue gas was remarkable. Especially benzo(a)pyrene was found mostly in the condensable fraction of the particles.

Keywords: biomass combustion, field measurement campaign, emission factors, benzo(a)pyrene, condensable particles.

1 INTRODUCTION

Domestic biomass heating appliances are very common in Austria and all over Europe. Thus, knowing their emissions in real life is important, since they contribute to air quality issues. Especially PM emissions from biomass combustion contributes significantly to PM pollution in Europe [1]–[3]. Moreover, focus on the polycyclic aromatic hydrocarbon benzo(a)pyrene (BaP) is given, due to its genotoxic and carcinogenic impact on human health [3], [4]. Real-life emission factors (EF) are used for emission inventories, air quality modelling or the prediction of air pollution impact on human health [5]–[7]. EF can be evaluated in two ways, by either close to real-life lab testing or by field measurements. One example of lab testing is the beReal test protocol for firewood room heaters [8]. During field measurements it was shown that this lab test method can reflect real-life situations [9]. Other lab tests showed a high variability of emission results at different close to real-life testing methods [10]–[13].

Close to real-life testing methods include transient conditions, like ignition, preheating or load changes of the heating appliances in order to reflect real-life conditions. Nevertheless, the broad variety of framework conditions (e.g. user influence, chimney design, heat output dimensioning etc.) cannot be considered in harmonized lab tests. Thus, field measurements are required in order to evaluate the broad range of emissions in real life.

In Austria, a comprehensive field measurement campaign was conducted by Spitzer et al. [14] in 1998. 173 biomass heating appliances were tested and average emission factors were evaluated. Within the project BioMaxEff [15], 16 newly installed biomass boilers were tested.
in the field at nominal load and varying load conditions close to real-life operation. EF need to reflect real-life conditions as close as possible. Thus, they have to be updated regularly, since the stock of biomass heating appliances is continuously changing.

In the project *Clean Air by biomass* a field measurement campaign was conducted measuring 15 different biomass heating appliances in the field at the end user. This study presents an overview of gaseous, particulate and BaP emission results and a comparison to current emission factors for Europe and Austria, respectively. Moreover, particle emissions and their chemical composition (BaP) in the hot and undiluted flue gas and in the cooled and diluted flue gas were compared.

2 MATERIAL AND METHODS

Within the *Clean Air by biomass* project a field measurement campaign was conducted, measuring 15 biomass heating appliances in total. These are:

- 6 room heaters (RH)
- 6 biomass boilers (BB)
- 3 tiled stoves (TS)

Table 1 gives an overview of the tested appliances. Their year of construction, nominal heat output, classification according to the respective EN standard and the evaluated emission parameters in the field are given. The appliances were installed in single family or farmhouses. Whereas boilers represented the main heating source, room heaters were used as additional heating source. Tiled stoves were used as both.

2.1 Testing procedure

During field measurements, all tested appliances were operated by the end user according to their usual operation habits. Fuel was also provided by the end user in order to reflect real-life conditions. Measurement of gaseous emissions, temperature and draught conditions of the flue gas was done continuously. Particulate emissions were measured discontinuously in each batch (RH and TS) or test phase (BB). Measurements lasted until the end of a heating cycle. For room heaters and tiled stove this was determined by the end user or at a maximum of three continuous batches. For boilers, three or four test phases were measured to evaluate different combustion phases – at least ignition and full load operation.

2.2 Measurement equipment and set up

For the field measurements two different equipment sets were used. Depending on the local situation in the field (mainly space demand and accessibility), the more extensive or the standard set was chosen (Table 2).

Measurements were done with a logging interval of 1 s for flue gas draught (p), temperature (T) and gaseous emissions, i.e. oxygen (O2), carbon dioxide (CO2), carbon monoxide (CO) and organic gaseous compounds (OGC). Particulate emissions (TSP and TSP40) were either measured according to the standard VDI 2066-1 (Standard set) or according to a new in-house developed TSP sampling procedure (Extensive set). Based on the method of Klauser et al. [16], it was adapted to enable simultaneous measurements of hot and cooled particles with only one suction device (it was necessary to have only one sampling point in the flue duct at the end user in the field). Thereby, a suction tube with a planefilter directly after the sampling nozzle and an additional planefilter after a diluter were used. The
Table 1: Overview of tested appliances.

<table>
<thead>
<tr>
<th>Appliance</th>
<th>Year of construction</th>
<th>Nominal heat output</th>
<th>EN standard</th>
<th>Evaluated parameters</th>
<th>Technology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Firewood room heater</td>
<td>2013</td>
<td>8 kW</td>
<td>EN 13240</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td>RH (n=6)</td>
</tr>
<tr>
<td>Firewood room heater</td>
<td>2017</td>
<td>8 kW</td>
<td>EN 13240</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td></td>
</tr>
<tr>
<td>Firewood room heater</td>
<td>~1997</td>
<td>8 kW</td>
<td>ÖNORM M 7520</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td></td>
</tr>
<tr>
<td>Firewood room heater</td>
<td>2013</td>
<td>7.3 kW</td>
<td>EN 13240</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td></td>
</tr>
<tr>
<td>Firewood insert</td>
<td>~2010</td>
<td>n.a.</td>
<td>EN 13229</td>
<td>CO, TSP</td>
<td></td>
</tr>
<tr>
<td>Firewood cooker</td>
<td>2016</td>
<td>20 kW</td>
<td>EN 12815</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td></td>
</tr>
<tr>
<td>Wood chip boiler</td>
<td>2001</td>
<td>40 kW</td>
<td>EN 303-5</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td>BB (n=6)</td>
</tr>
<tr>
<td>Wood chip boiler</td>
<td>2001</td>
<td>40 kW</td>
<td>EN 303-5</td>
<td>CO, TSP</td>
<td></td>
</tr>
<tr>
<td>Wood chip boiler</td>
<td>~1985</td>
<td>~60 kW</td>
<td>n.a.</td>
<td>CO, TSP</td>
<td></td>
</tr>
<tr>
<td>Boiler using saw dust</td>
<td>1962</td>
<td>250 kW</td>
<td>n.a.</td>
<td>CO, OGC, TSP, TSP40, BaP</td>
<td></td>
</tr>
<tr>
<td>Boiler using saw dust</td>
<td>2018</td>
<td>350 kW</td>
<td>EN 303-5</td>
<td>CO, OGC, TSP, TSP40</td>
<td></td>
</tr>
<tr>
<td>Firewood boiler</td>
<td>2010</td>
<td>35.6 kW</td>
<td>EN 303-5</td>
<td>CO, TSP</td>
<td></td>
</tr>
<tr>
<td>Firewood tiled stove</td>
<td>1985</td>
<td>n.a.</td>
<td>n.a.</td>
<td>CO, TSP</td>
<td></td>
</tr>
<tr>
<td>Firewood tiled stove</td>
<td>~1970</td>
<td>n.a.</td>
<td>n.a.</td>
<td>CO, OGC, TSP, TSP40</td>
<td></td>
</tr>
<tr>
<td>Firewood slow heat release appliance</td>
<td>2016</td>
<td>n.a.</td>
<td>EN 15250</td>
<td>CO, OGC, TSP, TSP40</td>
<td></td>
</tr>
</tbody>
</table>

n.a.: not available

Table 2: Overview of measured parameters with the two measurement equipment sets.

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Measured parameters</th>
<th>Number of measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extensive measurement set</td>
<td>CO, CO₂, O₂, OGC, T, p, TSP, TSP40, BaP</td>
<td>BB: n = 3 (4); TS: n = 2; RH: n = 5</td>
</tr>
<tr>
<td>Standard measurement set</td>
<td>CO, CO₂, O₂, T, p, TSP,</td>
<td>BB: n = 3 (2); TS: n = 1; RH: n = 1</td>
</tr>
</tbody>
</table>

First one is for the determination of particles in the hot flue gas (TSP) and was heated up continuously to 130°C. The second filter is for the determination of particles in cooled flue gas (<40°C), which includes condensable organic compounds (TSP40). The sum of both gives the total sampled particles (TSPtotal). Fig. 1 shows a scheme of the used suction device. After the measurements the filters were sent to an analyzing lab for the determination of benzo(a)pyrene (BaP) concentrations (Section 2.4).
2.3 Data evaluation

Data evaluation was done for each appliance for a whole heating cycle (Section 2.1). Thereby, for gaseous emissions (CO and OGC) a time weighted mean was calculated over the whole measurement period. Results are given in mg/m³ at standard temperature and pressure (STP), referred to 13% O₂ in dry flue gas. For particulate emissions (TSP and TSP40) results are firstly calculated for each measured batch/phase according to Klauser et al. [17]. The sum of TSP and TSP40 specifies the result for TSP_{total}. These results are given as well in mg/m³ at standard temperature and pressure (STP), referred to 13% O₂ in dry flue gas. Afterwards a time-weighted mean was calculated for the result of the whole heating cycle.

The proposed European and Austrian EF are given in mg/MJ. Thus, emission concentration results of the field measurements were transferred to mg/MJ based on combustion calculation [18] and according to the fuel composition. For the final evaluation results were aggregated according to the type of appliances. Means, medians, maxima and minima were calculated for the presentation of the results.

2.4 Chemical characterization of sampled particles

The TSP and TSP40 filters, taken during the field measurements, were characterized regarding BaP concentrations. This procedure was done according to the protocol of DIN EN 15549 using a solvent mixture of dichloromethane and cyclohexane for extraction. The chemical analysis itself was done with a GC-MS. Results are given in µg/m³. They are transferred to µg/MJ according to the fuel composition (same calculations as for other emissions – Section 2.3).
2.5 Fuel

Fuel for the field measurements was provided by the end user. To evaluate fuel properties, representative samples of the used fuel were taken and analyzed for carbon (C), hydrogen (H), nitrogen (N), water (w) and ash (a) content. The analyses were done according to the standards ISO 16948 (C, H, N), ISO 18134-2 (w) and ISO 18122 (a). The mean results are given in Table 3. All chemical parameters (C, H, N) of all tested fuels are in a narrow range, so the calorific values of the fuels are on a comparable level. The water content is in a range between 6 and 16 wt%. This means, that firewood as well as wood chips and saw dust are already dried. Especially for wood chips this indicates, that the fuel is provided by the user itself and stored in an appropriate way. The ash content is very low for all fuels.

Table 3: Overview of chemical analysis of fuels, used in the field measurements.

<table>
<thead>
<tr>
<th></th>
<th>C (wt%)</th>
<th>H (wt%)</th>
<th>N (wt%)</th>
<th>w (wt%)</th>
<th>a (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>49.5</td>
<td>6.1</td>
<td>0.1</td>
<td>10.7</td>
<td>0.7</td>
</tr>
</tbody>
</table>

3 RESULTS AND DISCUSSION

3.1 Emission results

Fig. 2 gives an overview of the emission results of the tested appliances, according to the type of technology. On average, room heaters have the highest emissions. This technology shows mean values of 4029 mg/MJ for CO, 712 mg/MJ for OGC, 138 mg/MJ for TSP and 226 mg/MJ for TSP total. The lowest gaseous emissions occur at boilers (CO: 914 mg/MJ, OGC: 56 mg/MJ). Particulate emissions were lowest at tiled stoves (TSP: 58 mg/MJ, TSP total: 67 mg/MJ). Medians (red cross) indicate that for room heaters and partly for boilers, a few higher values increase the mean values. In case of room heaters these values occurred with very bad user operation of the appliance (e.g. restricted air supply or overload batches). At boilers mainly the old boiler using saw dust as fuel had higher emissions.

Moreover, emissions have a high variability (which is common for emissions measured in the field), indicated by the whiskers (maximum and minimum) in Fig. 2. Especially room heaters have a wide range in emissions due to prevalent transient conditions during firewood burning and due to more possible influencing factors of the end user.

Figure 2: Overview of emission results (mean values) of the field measurement campaign. The whiskers indicate maximum and minimum values.
The study of Spitzer et al. [14] showed similar values for room heaters, but much higher values for boilers for CO (RH: 4463 mg/MJ, BB: 4303 mg/MJ) and OGC (RH: 664 mg/MJ, BB: 448 mg/MJ). TSP results are on a comparable level for both technologies (RH: 148 mg/MJ, BB: 90 mg/MJ). This indicates, that the stock of boilers changed to more advanced technologies with lower gaseous emissions.

The current field measurement results are comparable to the emissions of the study of Ozgen et al. [11]. There, lab tests with three consecutive batches at a firewood stove including the ignition and preheating batch were conducted. The measured emissions showed values of 3196 mg/MJ for CO and 110 mg/MJ for TSP.

Another study from Pettersson et al. [12] presents results from lab tests with two full load batches at a firewood stove. This study also takes maloperation into account, i.e. restricted air supply and moist wood. Emissions are 3600 mg/MJ for CO, 820 mg/MJ for OGC and 140 mg/MJ for TSP total. OGC and TSP total are higher than the presented values of the field measurements and of the study of Ozgen et al. [11]. This reveal that unfavorable user habits can increase those emissions.

The results of the field measurements in the BioMaxEff [15] project showed lower emissions for boilers on average (CO: 243 mg/MJ, OGC: 5 mg/MJ, TSP: 15 mg/MJ). However, these results only include measurements at new installed pellet boilers, which were not tested in the current study.

3.1.1 BaP emissions
As illustrated in Fig. 2 it is even more obvious, that BaP emissions are highest for room heaters in the field. This technology shows an average value of 305 µg/MJ compared to 18 µg/MJ for boilers and 4 µg/MJ for tiled stoves. Also, the range of BaP emissions in the field is very high. Results indicate that the user could be a major influencing factor on BaP formation. However, further investigations are needed to evaluate most important influencing factors of the formation of BaP.

The study of Klauser et al. [10] showed clearly lower values for BaP with a maximum of 86 µg/MJ for firewood appliances. However, only new and advanced technologies were tested. Moreover, the operation of the appliance at beReal testing follows the manual of the manufacturer, so maloperation is omitted.

Ozgen et al. [11] found BaP emissions of 204 µg/MJ which is on average lower, but in the range of the current study. Pettersson et al. [12] found higher values (BaP: 610 µg/MJ) at restricted air supply and the use of moist wood for testing. Compared to the current study this is at the level of the maximum value of the measurements (646 µg/MJ).

Klauser et al. [16] measured average BaP emissions at a state-of-the-art wood chip boiler of 3.5 µg/MJ at starting conditions. Since, these conditions are normally more likely to formulate BaP emissions, the result is clearly lower compared to the emissions of the current study.

3.2 Comparison to current emission factors for Europe and Austria

Fig. 3 shows the single emission results for OGC, BaP, TSP and TSP total of the tested appliances in comparison to currently suggested emission factors for Europe (EMEP) and Austria (AEF). In Table 4, the different emission factors for Europe and Austria, which were used for comparison, are given.

The comparison of the EF of the field measurements (Fig. 2 and Fig. 3) and the suggested EF (Table 4 and Fig. 3), shows that EF for boilers measured in this study are much lower than the proposed European EF, when using EMEP_d (conventional boilers), except for 1
TSP result. This outlier is the result of the old boiler fired with saw dust. The 95% confidence interval given for European EF, cover this higher value. The lowest results of the field measurement campaign are well reflected by EMEP_c (pellet boilers), although other technologies were tested, except for particulates which are higher by trend. EMEP_c (advanced/eco-labelled boilers) show higher EF for gaseous emissions (CO, OGC) and represents particulates and BaP, compared to the field measurements. Tiled stoves are compared with EMEP_b (high-efficiency stove) and EMEP_c (advanced/eco-labelled stove). EMEP_b shows higher EF than the emissions of tiled stoves of this study. EMEP_c would represent the emissions of tiled stoves, except for OGC the proposed EF is higher. EMEP_a (conventional stove), EMEP_b (high efficiency stove) and EMEP_c (advanced/eco-labelled stove) are used for comparison with room heaters. The range of those EF comprises the values of the field emissions well, with exception of some cases. For BaP 3 clearly higher results are measured. Nevertheless, they are within the 95% confidence interval of EMEP_a and EMEP_b.

Figure 3: Emission factor results for OGC, BaP, TSP and TSP_total of the field measurements (EF_field) in comparison to current suggested European (EMEP) and Austrian (AEF) emission factors (EF_sugg), differentiated by technology (BB: boilers, TS: tiled stoves, RH: room heaters).
Table 4: Overview of emission factors for Europe [5] and Austria [6] in mg/MJ.

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
<th>CO</th>
<th>OGC</th>
<th>TSP</th>
<th>TSP$_{total}$</th>
<th>BaP</th>
</tr>
</thead>
<tbody>
<tr>
<td>EMEP_a</td>
<td>Conventional stoves</td>
<td>4000</td>
<td>750*</td>
<td>200</td>
<td>800</td>
<td>121</td>
</tr>
<tr>
<td>EMEP_b</td>
<td>High-efficiency stoves</td>
<td>4000</td>
<td>437.5*</td>
<td>170</td>
<td>400</td>
<td>121</td>
</tr>
<tr>
<td>EMEP_c</td>
<td>Advanced/eco-labelled stoves and boilers</td>
<td>2000</td>
<td>312.5*</td>
<td>54</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>EMEP_d</td>
<td>Conventional boilers &lt;50kWth</td>
<td>4000</td>
<td>437.5*</td>
<td>170</td>
<td>500</td>
<td>121</td>
</tr>
<tr>
<td>EMEP_e</td>
<td>Pellet stoves and boilers</td>
<td>300</td>
<td>12.5*</td>
<td>32</td>
<td>62</td>
<td>10</td>
</tr>
<tr>
<td>AEF_i</td>
<td>Wood stoves and cooking stoves</td>
<td>3955.9</td>
<td>736.35*</td>
<td>148</td>
<td></td>
<td>86.25**</td>
</tr>
<tr>
<td>AEF_ii</td>
<td>Tiled wood stoves and masonry heaters</td>
<td>2345.3</td>
<td>422.5*</td>
<td>100</td>
<td></td>
<td>42.5**</td>
</tr>
<tr>
<td>AEF_iii</td>
<td>Natural-draft wood boilers</td>
<td>3483</td>
<td>437.5*</td>
<td>75</td>
<td></td>
<td>8.75**</td>
</tr>
<tr>
<td>AEF_iv</td>
<td>Forced-draft wood boilers</td>
<td>3234.2</td>
<td>406.25*</td>
<td>50</td>
<td></td>
<td>0.5**</td>
</tr>
<tr>
<td>AEF_v</td>
<td>Wood chips boilers with conventional technology</td>
<td>2400</td>
<td>540.5*</td>
<td>100</td>
<td></td>
<td>6**</td>
</tr>
<tr>
<td>AEF_vi</td>
<td>New wood stoves and cooking stoves</td>
<td>2345.3</td>
<td>453.61*</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*EF are given for NMVOC; based on Anderl et al. 2017 for biomass combustion a methane share of 25% is in OGC; EFOGC = EF$_{NMVOC}$ + 0.25 EF$_{NMVOC}$.

**Austrian EF are only given for the sum of 4 PAHs [6]; the share of BaP is given as 25%; EFBaP = 0.25 EFPAH.

Austrian EF for boilers are in general too high for gaseous emissions (CO and OGC) in comparison to the study results. Particulates are well represented by AEF_iii (natural-draft wood boiler), AEF_iv (forced-draft wood boiler) and AEF_v (wood chips boiler with conventional technology). There is one EF for tiled stoves (AEF_ii...tiled wood stoves and masonry heaters). This EF overestimates emissions in comparison to the conducted field measurements of this study. For all room heaters, AEF_i (wood stoves and cooking stoves) represents a reliable mean value. Some emissions are clearly higher, others are clearly lower than this EF. AEF_vi (new wood stoves and cooking stoves) represents the lower values of this study.

Especially for room heaters it is very hard to define appropriate EF even when distinguishing between different technologies, since the variability is very high. Moreover, the operation of room heaters is influenced by many factors regarding the framework, foremost user operation habits. The availability of a 95% confidence interval like it is available for European EF, is a good measure for modeling best or worst-case scenarios of air quality in a region. Nevertheless, this wide range of emissions in the field makes it hard to predict air quality or conduct emission inventories.
3.3 Differences of particulate measurements

For the field measurement, tested with the extensive equipment, TSP and TSP40 samples are compared (Fig. 4). Thus, an evaluation of the share of condensable particles is possible. Moreover, the fraction of BaP in the condensable particles is determined. Results show that a high share of TSP\textsubscript{total} is in the hot flue gas (TSP). However, for room heaters 33\% can be found in the condensable fraction (TSP40). Moreover, emissions of BaP are mainly found in the condensable fraction. For air quality issues the sum of both fractions is fundamental, since a fraction of volatile organic compounds condensates at ambient temperatures. Thus, measuring this sum of particles is necessary to get reliable emission factors for TSP\textsubscript{total}. Otherwise, emissions could be underestimated. This is even more important regarding BaP and the impact of air quality on human health.

![Figure 4: Comparison of particle measurement and the chemical characterisation in hot (dark) and cooled (bright) flue gas.](image)

4 CONCLUSION

In this study results of a field measurement campaign, including 15 biomass heating appliances are presented. The following conclusions can be drawn:

The results of this study in comparison to the study of Spitzer et al. indicate that for boilers a technological development towards lower gaseous emissions had happen. For room heaters this was not found, even though the results of official type tests assume such a development. However, room heaters are more influenced by the end user. End user training and also focus on the optimization of real-life conditions during heating (including ignition, preheating and load changes, which is not tested at official type tests) could have a high potential for future improvement.

The proposed European and Austrian EF show higher values by trend for boilers and tiled stoves. For room heaters EF are in the range of emissions of this study. Combined with the comparison to the results of Spitzer et al. of 1998, this reveals that EF, especially for boilers and tiled stoves, need to be updated regularly. Moreover, due to the high variability of the field measurement results it is very hard to define EF. This high variability is mainly caused by framework conditions (user influence, installation in the house, dimensioning, chimney...).
Close to real-life lab tests can be a good opportunity to reflect the technological development of heating appliances at close to real-life conditions. Nevertheless, they cannot reflect the whole framework. Since EF are used for emission inventories, they have an impact on policy decision makers and regulations. Thus, field measurements should be fostered in order to get a good overview of the emissions in real life and even more reliable EF.

The comparison of particles in the hot and cooled flue gas reveal that the amount of condensable particles is unneglectable. Moreover, BaP is predominantly found in the condensable fraction of particles. Hence, EF should consider condensable particles. Otherwise TSP_{total} and BaP can be underestimated.

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