Determining PM-emission fractions (PM_{10}, $PM_{2.5}$, $PM_{1.0}$) from small-scale combustion units and domestic stoves using different types of fuels including bio fuels like wood pellets and energy grain

Ehrlich, Chr., Noll, G., Kalkoff, W.D. Saxony-Anhalt Environment Agency (Landesamt für Umweltschutz Sachsen- Anhalt), D-06009 Halle (Saale), PF 200841, Germany, Telephone: +49 345 5704 500, Fax: +49 345 5704 505, e-mail: ehrlich@lau.mlu.lsa-net.de

1 Abstract

Background: The European Commission's "Thematic Strategy on Air Pollution" (Communication, 2005), which was developed as a long-term, strategic and integrated policy advice to protect against significant negative effects of air pollution on human health and the environment, addresses small scale combustion units as increasingly important emission sources. In this context emission measurement programmes were carried out at small-scale combustion units and domestic stoves in Germany (6 kW - 450 kW) to determine the fine dust in waste gases.

Methods: The PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ fractions in the waste gas were sampled using a cascade impactor technique. Additional total dust concentrations were determined. Waste gas samples were taken from small-scale combustion units and domestic stoves, firing lignite briquettes, wood, wood pellets and energy grain.

Results: In total 250 individual measurement results were obtained during 31 different measurement campaigns. The study found out that the PM_{10}-proportions for all results vary typically between 92 - 99 % of the total dust, the $PM_{2.5}$-proportion between 79 - 97 % and the $PM_{1.0}$ proportion between 63 - 91 % of the total dust.
Typical particle size distributions for different combustion processes, when presented as cumulative frequency distributions, were nearly straight lines when plotted as a RRSB diagram. For more structural information the particle size distributions were also presented as logarithmic frequency distributions. The particle size distributions determined for the different combustion units and different fuels show interesting similarities with high to very high portions of fine particles. In comparison to other industrial processes like mechanical, chemical or mixed processes clear differences could be identified. In these cases the particle size distributions show obviously coarser particle proportions.

Conclusion: The results demonstrate that solid fuel combustion in small-scale units without abatement technique induced enhanced levels of PM-emission and very high proportions of fine particles in the waste gas. The highest levels of total dust emission concentration comparing our bio fuel results were found, if energy grain was used as fuel. Wood pellet combustion produced the lowest total dust emission.

Keywords: small scale combustion units, PM_{10}-, $PM_{2.5}$-, $PM_{1.0}$-emission, bio fuel, wood pellets, energy grain, impactor, emission measurement

2 Introductions

The European Commission's "Thematic Strategy on Air Pollution" (Communication, 2005), which was developed as a long-term, strategic and integrated policy advice to protect against significant negative effects of air pollution on human health and the environment (WHO Air quality guidelines, 2006), addresses small scale combustion units as increasingly important emission sources.

On the other hand the European Commission actively supports the use of biomass for producing energy. The aim is to increase the use of renewable energy and to avoid an increase of the greenhouse gas CO_2 in the global atmosphere. Partly because of this support the use of wood and non-wood biomass fuels is increasing across the EU member states. Small-scale biomass combustion is normally used for heat production. Biomass is often used in small-scale combustion units. Low costs associated with short transportation distances have increased the interest in using various bio fuels in small-scale heating plants. Using non-fossil fuels has numerous advantages. However, some of the emissions produced can still be harmful e.g. soot, fine particulates, hydrocarbons and carbon monoxide and NO_x. Small-scale biomass combustion has been proven to be a very important cause of fine particle emissions together with emissions from the transportation sector. This problem is recognized by authorities in the EU-countries and new and more stringent regulations limiting the emissions are expected to become implemented in the near future. At present there are no common international standards for emission control from small-scale bio fuel burning, national or even regional emission standards are used.

In this context emission measurement programmes were carried out at small-scale combustion units and domestic stoves in Germany (6 - 450 kW) to determine the fine dust in waste gases.

3 Experimental

3.1 Sampling

The PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ fractions in the waste gas were sampled (VDI 2066 Bl. 5, 1994) using cascade impactors as 8- or 6-stage Anderson Impactors, type Mark III (material: stainless steel) and/or with a 6-stage impactor of the company Stroehlein type STF 1 (material: Titanium). Preliminary investigations showed that technology-based particle size distributions determined with both impactors agreed well under same sampling conditions. The impactor measurements had normally been performed as grid measurements in accordance with the guideline (VDI 2066 Bl. 1, 2006, EN 13284-1).
Additional total dust concentrations were determined to be able to specify the required suction times for the impactor samplings. For the particle separation perforated collecting plates and the backup filter made of glass fibre material were used. The determination of the sucking sample gas flow was carried out by means of thermal mass flow meters.
The high dust concentrations at the investigated domestic fire places often required sampling times, substantially shorter than the necessary time for the complete burn-up of the fuel. However, investigations proved that in the first third of the combustion cycle (in this time the impactor measurements took place) most of the particle load had been emitted.
Waste gas samples were taken from small-scale combustion units and domestic stoves, firing lignite briquettes, wood, wood pellets and energy grain.
The operating conditions of the respective stove or combustion unit were determined and documented during the sampling periods. Furthermore other relevant pollutants or components, e.g. CO, CO_2, NO_x, O_2 and SO_2 in the waste gas of the plant were

measured with standardized measurement methods (VDI guidelines, CEN standards) and officially approved measurement instruments. Also the flue gas conditions (velocity, static pressure, temperature and water vapour content) were determined. As an example the test arrangement and the installation of measurement ports on the household stove (table1, No 1 - 8) are shown in figure 1.

Fig. 1: Test arrangement household stove (brown coal briquette)

All measurements were performed with a set of quality assurance measures according to (EN ISO/IEC 17025, 2005). All combustion operating and measured parameters were documented carefully according to a standardized report guideline

3.2 Small scale combustion units and domestic stoves

The tested small-scale combustion units and domestic stoves are described by their performance characteristics and used fuels in Table 1. Also the numbers of PMx-samplings and the determined values of total dust, PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ as well as the kind of dust separation are given.
The investigated small-scale combustion units and domestic stoves can be divided in four groups: combustion of coal (brown coal briquette), wood, energy grain and wood pellets. Stoves and small scale firing units were tested in the range of performance of 4 kW to 420 kW.

4 Results and Discussion

In total 250 individual measurement results were obtained during 31 different measurement campaigns (Universität Stuttgart 1999, 2001; Landesamt für Umweltschutz 1997 – 2006; Ehrlich et. al. 1999, 2000).

Table 1: List of investigated plants with performance characteristics, assigned fuels and measurement results

		coal							
1		LAUBAG-bcb[1)2)]	3	6 kW	without				
2		LAUBAG-bcb[1)3)]	2	6 kW	without				
3		MIBRAG-bcb[1)2)]	3	6 kW	without				
4		MIBRAG-bcb[1)3)]	3	6 kW	without				
5		Polish bcb[1)2)]	2	6 kW	without				
6		Polish bcb[1)3)]	2	6 kW	without				
7		Bashkirian bcb[1)]	3	6 kW	without				
8		MIBRAG-bcb[1)]	3	6 kW	without				
		wood							
9		chips	1	177 kW	cyclone				
10		chip board	1	139 kW	cyclone				
11		chips	1	148 kW	without				
12		chips	1	43.4 kW	without				
13		joinery residues	1	133 kW	without				
14		coloured pencil residues	1	112.5 kW	without				
15		log wood	1	416.5 kW	multi-cyclone				
16		log wood	1	273 kW	multi-cyclone				
17		log wood beech	1	9.4 kW	without				
18		log wood beech	1	7.5 kW	without				
19		log wood pine	1	8.2 kW	without				
20		log wood pine	1	6.8 kW	without				
21		log wood beech	1	5.7 kW	without				
22		log wood beech	1	4.1 kW	without				
		energy grain							
23		winter barley	3	38 kW	without				
24		winter barley	3	21 kW	without				
25		winter wheat	3	41 kW	without				
26		winter wheat	3	22 kW	without				
27		winter barley	3	39 kW	without				
28		winter barley	3	21 kW	without				
		wood pellets							
29		wood pellets	1	8.0 kW	without				
30		wood pellets	3	39 kW	without				
31		wood pellets	3	22 kW	without				

1) bcb = brown coal briquette
2) after cooling
5) n.m. = no measurement
3) before cooling
4) standard temperature (273.15 K), pressure (101.3 kPa) after correction for the water vapour content, O_2- content 11%

It is common practice to plot size distribution data in such a way that a straight line is the result. This simplifies data analysis, e.g. for reading of essential parameters like PM_{10}, $PM_{2.5}$ or $PM_{1.0}$ and can be done if the distribution fits a standard law. In this study the best fit with the experimental data was gained by using the cumulative frequency particle size distribution (D) according to Rosin, Rammler, Sperling, and Bennet (RRSB) (Batel 1964). Another illustration method is the depiction of the frequency distribution (Y) over the PM diameter, which has been used too:
$Y = -dR/d(\log d)$ (R is the cumulative residue distribution, $R = 1 - D$) (Batel 1964).

The particle size distributions determined for the different combustion units and different fuels show interesting similarities with high to very high proportions of fine particles. In comparison to other industrial processes like mechanical, chemical or mixed processes clear differences could be identified. In these cases the particle size distributions show obviously coarser particle proportions (Ehrlich, C. et. al., 2006).
Figs. 2 - 9 typify the particle size distributions in the flue gases of small combustion plants.

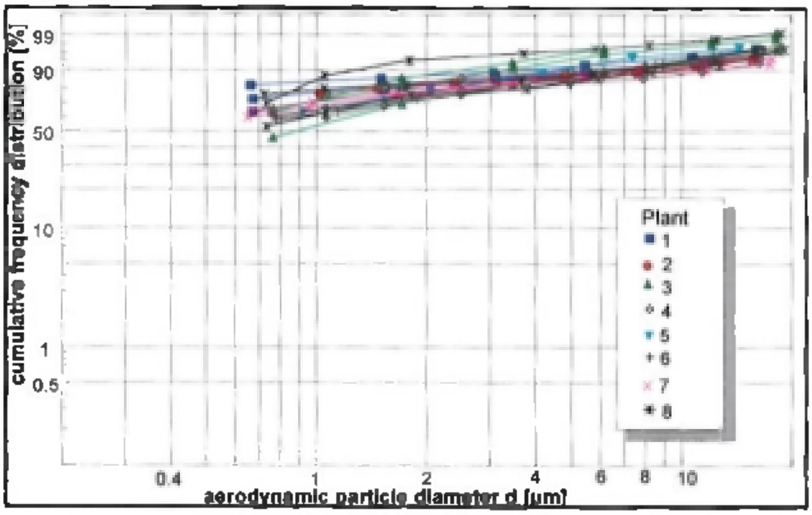

Fig. 2: PM size distributions of brown coal briquette firing; RRSB cumulative frequency distribution (numbers according to table 1)

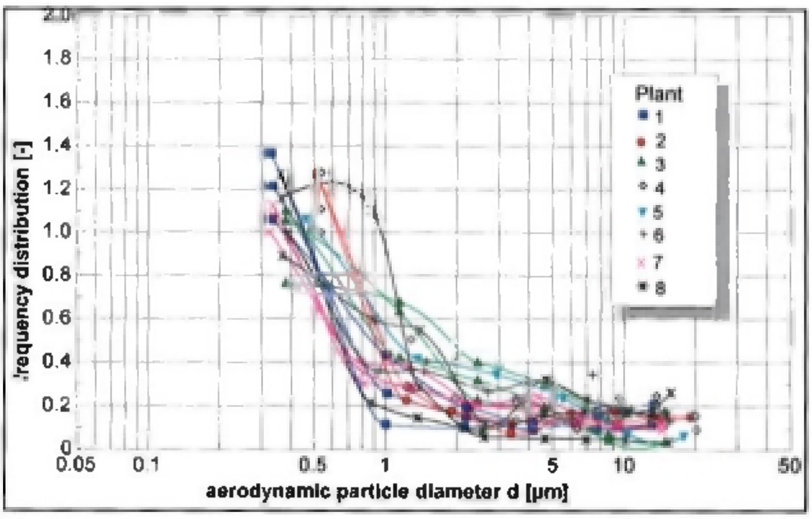

Fig. 3: PM size distributions of brown coal briquette firing; frequency distribution (numbers according to table 1)

Fig. 2 and 3 show the particle size distributions of the emissions from various types of brown coal briquette fired in a household stove. In general a bi-modal particle size distribution appears in the combustion processes (Szpila et. al. 2003). The density distribution in Fig. 3 shows two modes, one in the submicrometer range (0.3 - 0.8 µm) and the other (smaller) in the coarse range (5 - 10 µm).

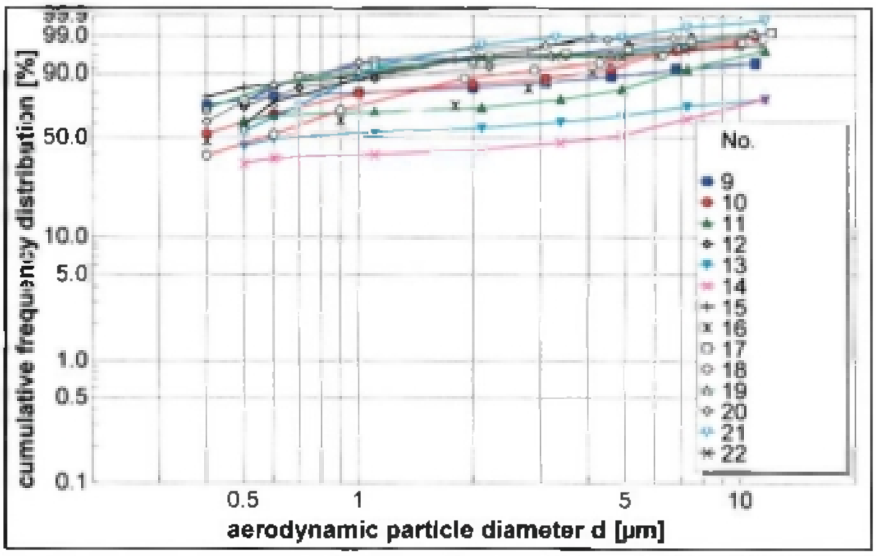

Fig. 4: PM size distributions of wood firing; RRSB cumulative frequency distribution (numbers according to table 1)

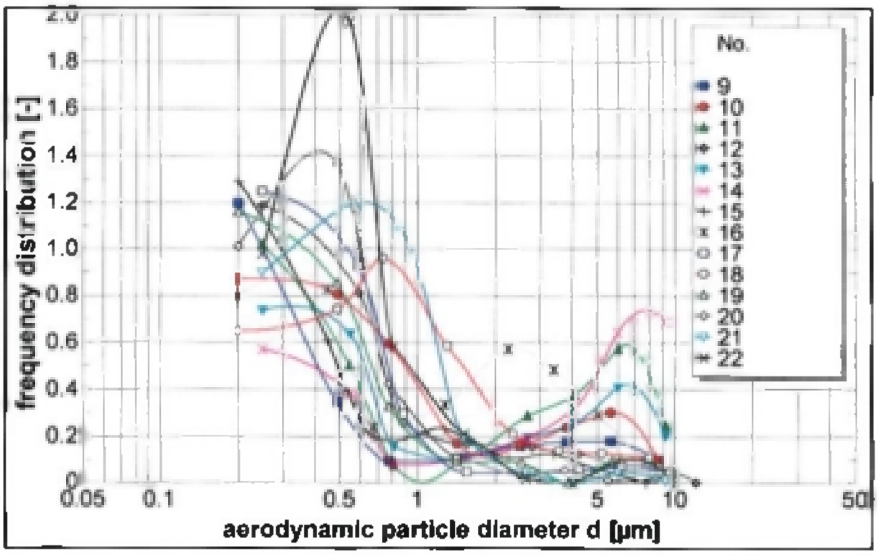

Fig. 5: PM size distributions of wood firing; frequency distribution (numbers according to table 1)

The particle size distributions of the 9 kW furnaces fired with log wood (beech, pine) (see plants No. 17 - 22 in Figs. 4 and 5) showed a greater proportion of fine particulates (see also table 1) than briquette-fired plants of a similar capacity (plants No. 1 to 8 in Figs. 2 and 3). The fine particulate fraction, which extends into the ultrafine range, can not be completely collected by the measuring technique used here (VDI 2066 Bl. 5 1994). These fine and ultrafine particles are formed from volatile exhaust components that condense during cooling in the exhaust duct and deposit on the surfaces of existing fine particles (such as soot) or form new particles. In the case of biomass combustion, in this case wood, volatile alkali compounds such as potassium salts are also present. Wood combustion plants with larger capacity (175 - 450 kW, plant No. 9 - 16, Figs 4 and 5) show distinctly coarser PM distributions. Here, it seems that coarser mineral ash components are carried away by highflow caused by the suction ventilators thereby enhancing the "coarse" mode.

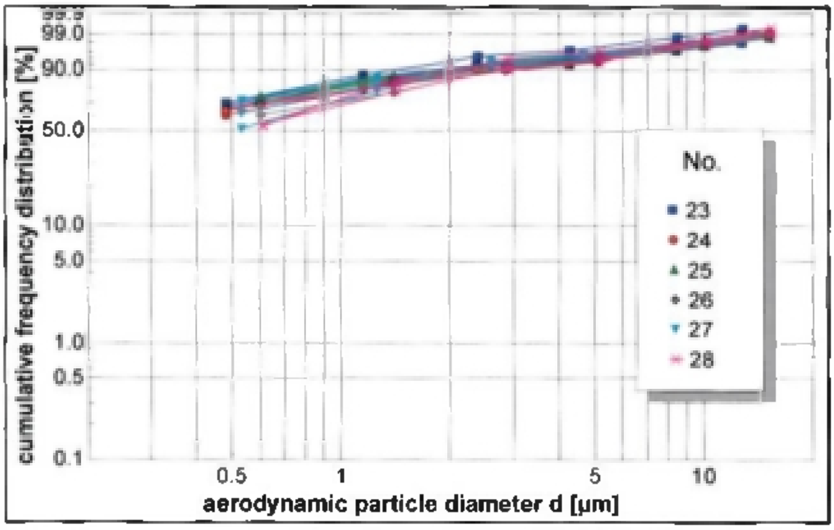

Fig. 6: PM size distributions of energy grain firing; RRSB cumulative frequency distribution (numbers according to table 1)

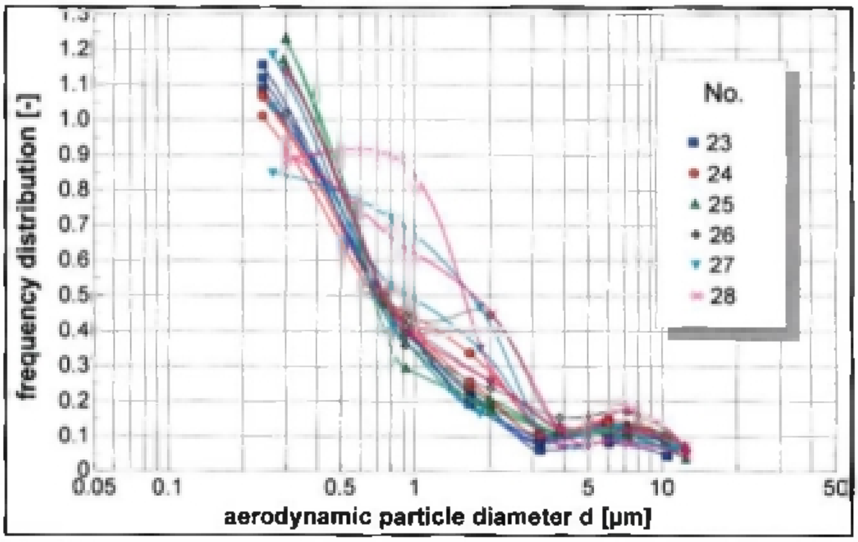

Fig. 7: PM size distributions of energy grain firing; frequency distribution (numbers according to table 1)

Firing energy grain shows similar fine PM size distributions. Similar to the wood combustion fine and ultrafine particles are formed by volatile exhaust components like potassium salts that condense during cooling in the exhaust duct and deposit on the surfaces of existing fine particles (such as soot) or form new particles. In (Nussbaumer, T., 2003) it was shown, that particles from wood combustion are mainly formed by nucleation, coagulation, and condensation during temperature decrease in the boiler.

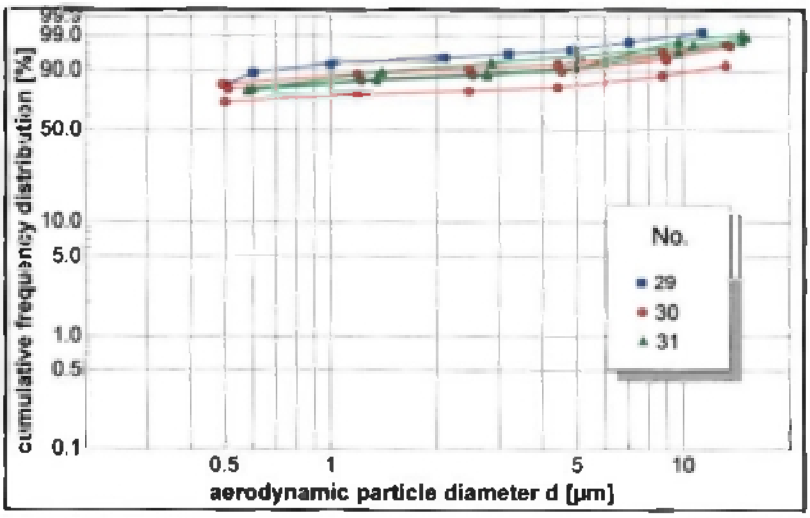

Fig. 8: PM size distributions of wood pellets firing; RRSB cumulative frequency distribution (numbers according to table 1)

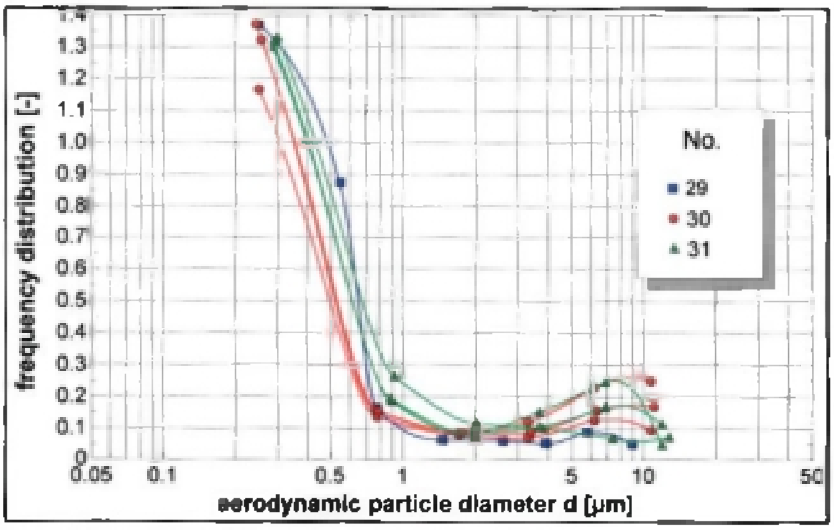

Fig. 9: PM size distributions of wood pellets firing; frequency distribution (numbers according to table 1)

Firing wood pellets shows very fine PM size distributions comparable with the PM size distribution of emitted particles from wood combustion stoves (4 - 10 kW).

Further evaluations were made of the individual plant groups[1] investigated. The respective PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ values are represented in box whisker diagrams – see Figs. 10, 11 and 12. A box indicates the range of the 25 and 75 percentile of the measured values and the biggest and smallest value of the measurement are indicated with a cross, a (-) indicates the 50 percentile.

[1] The results of plant No 13 and 14 (table 1) are not included in the following data analysis, since in these cases residues or waste materials and not untreated wood were combusted.

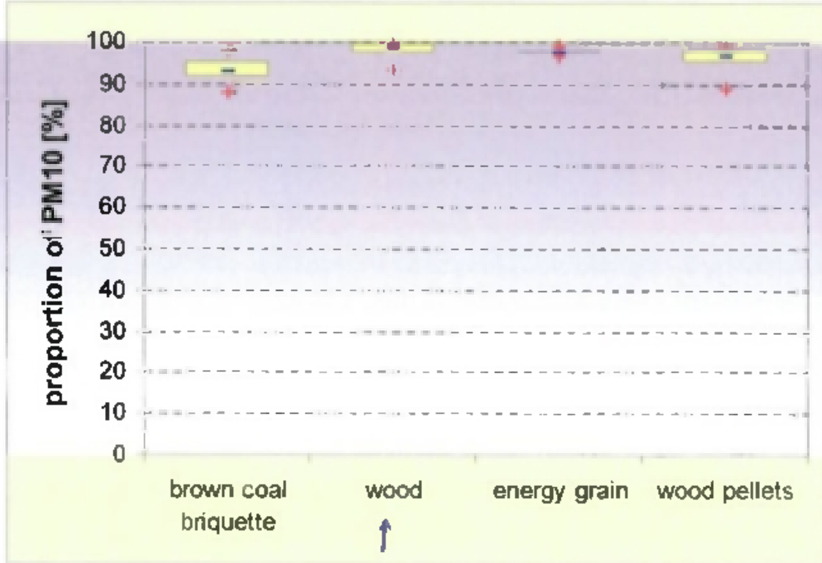

Fig. 10: Proportion of PM_{10} in percent (%) for all investigated small scale combustion units

In the study was found out that the PM_{10}-proportions for all results vary typically between 92 and 99 % of the total dust.

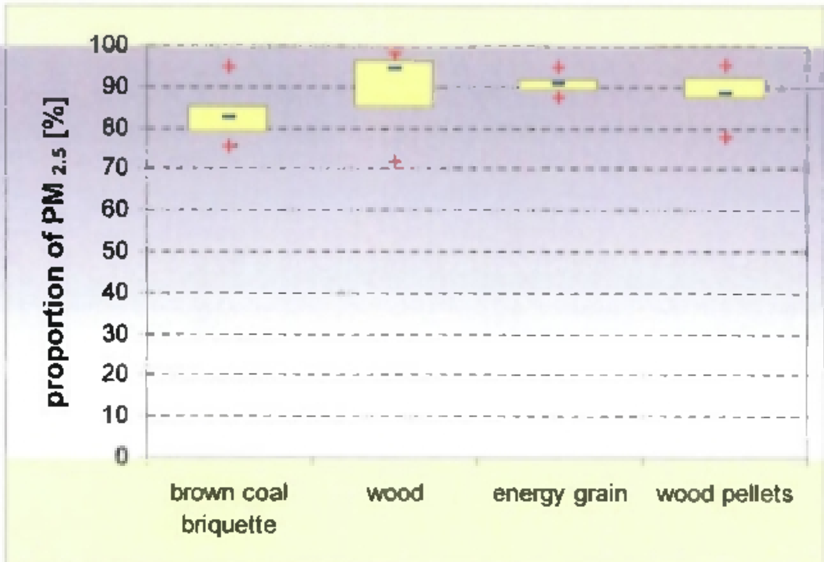

Fig. 11: Proportion of $PM_{2.5}$ in percent (%) for all investigated small scale combustion units

The $PM_{2.5}$-proportion of all results ranges between 79 - 97 %.

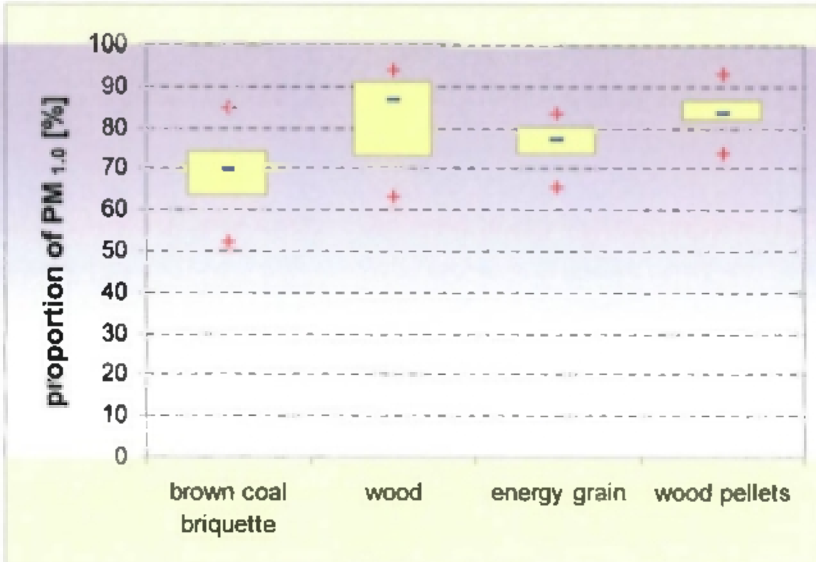

Fig. 12: Proportion of $PM_{1.0}$ in percent (%) for all investigated small scale combustion units

The $PM_{1.0}$ proportion of all results varies between 63 - 91 % of the mass concentration of the emitted total dust.

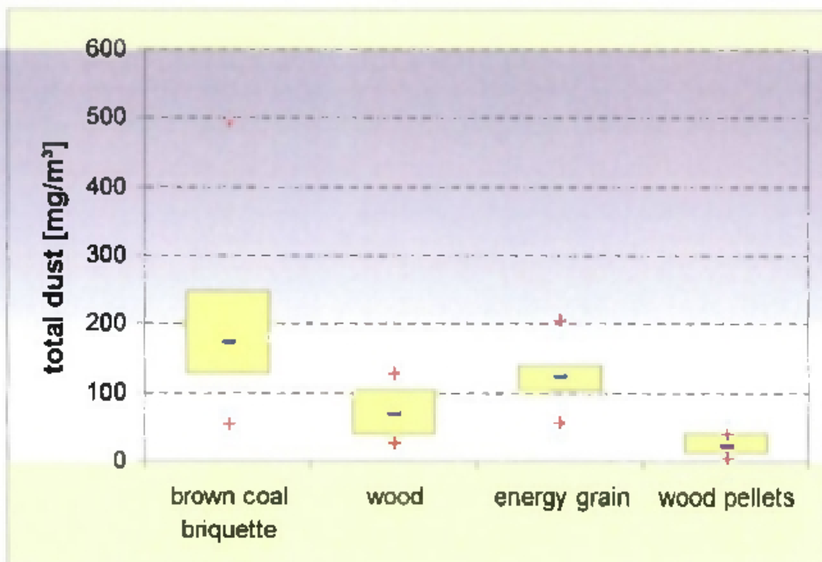

Fig. 13: Total dust (PM) in mg/m³ for all investigated small scale combustion units (mg/m³ related to standard temperature (273.15 K), pressure (101.3 kPa) after correction for the water vapour content, O_2 content 11%)

In comparison the results show enhanced total dust emission concentration, firing coal or bio fuels in small scale combustion units without abatement technique. Firing energy grain, our results range from 100 to 140 mg/m³ of total PM, on the other hand firing of wood pellets leads to 10 - 40 mg/m³.

This corresponds in principle with literature (Nussbaumer, T. 1999, 2003): Biomass combustion leads to relatively high emissions of particulates, i.e., well above 50 mg/m³ at 11 Vol. % O_2. The majority of the particulates are smaller than 10 µm (i.e., particulate matter PM_{10}) with a high share of submicron particles ($PM_{1.0}$).

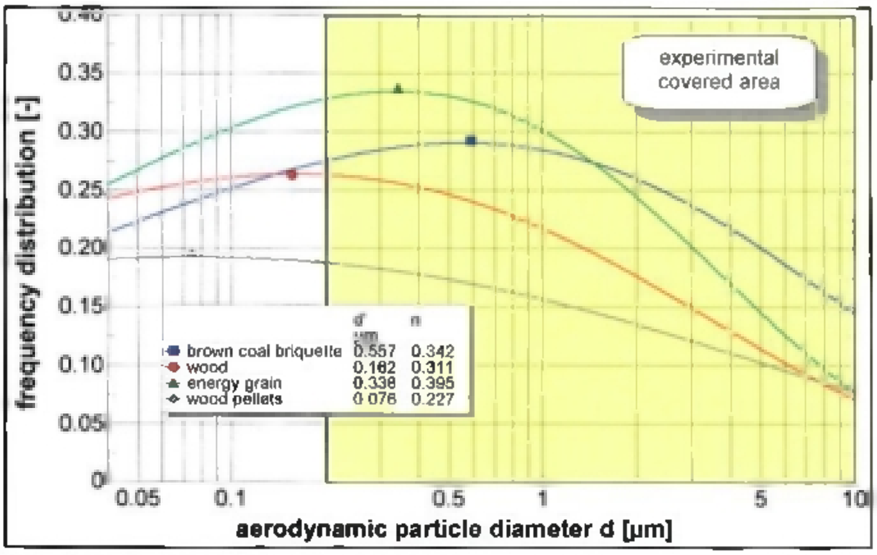

Fig. 14: Idealized and averaged PM size distribution as frequency distribution for all investigated small scale combustion units (d' and n according to RRSB, (Batel, 1964))

Considering the idealized and averaged PM size distributions (Fig. 14) of all investigated coal and biomass small scale combustion unit groups it is obvious, that the parameter d' (most frequent particle diameter) for wood pellet boilers lies in the range of the ultrafine particles (0.07 - 0.1 µm) and for brown coal briquette stoves, wood boilers and energy grain boilers in the submicron range (0.1 - 0.6 µm).

5 Conclusions

The results demonstrate that solid fuel combustion in small-scale units without abatement technique induced enhanced levels of PM-emission and very high proportions of fine particles in the waste gas. The highest levels of total dust emission concentration comparing our bio fuel results were found, if energy grain was used as fuel. Wood pellet combustion produced the lowest total dust emission.

6 Acknowledgements

The authors want to thank Birk Maiwald, Arnfried Reifert, Andreas Pohle, Marlies Hennig for the measurements in Saxony-Anhalt and the practical work, Professor Günter Baumbach and Anja Dreiseidler from the University of Stuttgart for their measurement results and Johannes Rumpler from the Saxony-Anhalt Agency for Agriculture, Forest and Horticulture for assistance with the measurements.

7 References

Batel, W.: Einführung in die Korngrößenmesstechnik, Springer-Verlag 1964

Communication from the Commission to the Council and the European Parliament, Thematic Strategy on air pollution, (COM (2005) 446)

Ehrlich, C., Noll, G., Kalkoff, W.-D.: Emissionsuntersuchungen von PM_{10}, $PM_{2.5}$ und $PM_{1.0}$ an Industrieanlagen und Hausbrandfeuerstätten. Zeitschrift Immissionsschutz, Berlin, 4 (2000) pp. 141 –148

Ehrlich, C., Noll, G., Kalkoff, W.-D.: Messtechnische Ermittlung von PM_{10}-und $PM_{2.5}$-Emissionen aus Industrieanlagen und Hausbrandfeuerstätten. Neuere Entwicklungen bei der Messung und Beurteilung der Luftqualität, Deutschland, Heidelberg, 27. bis 29. April 1999. VDI-Verlag Düsseldorf, VDI-Berichte: Nr. 1443. pp. 117-129

Ehrlich, C., Noll, G., Kalkoff, W.-D, Baumbach, G., Dreiseidler, A.: PM10, PM2,5 und PM1,0-Emissionen aus Anlagen – Ergebnisse von Messprogrammen des Bundes und der Länder, in : Feinstaub und Stickstoffdioxid, Beuth-Verlag, Berlin 2006, pp.129 -159

EN ISO/IEC 17025:2005, General requirements for the competence of testing and calibration laboratories

Landesamt für Umweltschutz Sachsen-Anhalt, Emissionsermittlungs-Berichte 01-97, 02-97, 02-99, 03-99, 01-00, 02-01, 03-01, 01-05, 02-05, 02-06, 03-06, unveröffentlicht

Landesamtes für Umweltschutz Sachsen-Anhalt, Feinstaubemissionsuntersuchungen in Sachsen-Anhalt: PM_{10}-, $PM_{2.5}$- und $PM_{1.0}$ – Emissionen aus Industrie und Hausbrand, Sonderheft 1/2001

Nussbaumer, T.; Hasler, P. Bildung und Eigenschaften von Aerosolen aus Holzfeuerungen. Holz als Roh- Werkstoff 1999, 57, 13 - 22.

Nussbaumer, T. Combustion and Co-combustion of Biomass: Fundamentals, Technologies, and Primary Measures for Emission Reduction. Energy & Fuels 2003, 17, 1510-1521

Szpila, A., Strand, M., Pagels, J., Lillieblad, L., Rissler, J. Gharibi, A., Bohgard, M., Swietlicki, E., Sanati, M., Particle Emissions from Biomass Combustion, Växjo University, Lund University, Sweden 2003

Universität Stuttgart, Institut für Verfahrenstechnik und Dampfkesselwesen, Feinstaubuntersuchungen an Holzfeuerungen, Bericht Nr. 44-1999

Universität Stuttgart, Institut für Verfahrenstechnik und Dampfkesselwesen, Korngrößenverteilung (PM_{10} und $PM_{2.5}$) von Staubemissionen relevanter stationärer Quellen, Abschlussbericht zum UBA Forschungsvorhaben 298 44 280-2001

VDI 2066 Bl. 1 Particulate matter measurement; measuring of particulate matter in flowing gases; gravimetric determination of dust load; fundamentals, 2006

VDI 2066 Bl. 5 Particulate matter measurement - Dust measurement in flowing gases; particle size selective measurement by impaction method - Cascade impactor,1994

WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide
Global update 2005 Summary of risk assessment, World Health Organization 2006, Geneva