

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

**CHARACTERISATION OF PARTICLE EMISSIONS FROM
SMALL-SCALE BIOMASS COMBUSTION**

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ABSTRACT

Several studies have shown that particulate air pollution is associated with increased mortality. Combustion devices like boilers and internal combustion engines are important sources of particles in the ambient air. In Sweden, the combustion of biomass is increasing and may consequently lead to locally increased particle concentrations. Roughly spoken, particles from biomass combustion consist of ash and of particles originating from incomplete combustion. Several studies of emissions of particles from biomass combustion have been performed, but the particle formation mechanisms are not yet fully understood. The purpose of the present work is to characterise particle emissions from small-scale combustion of wood pellets and wood briquettes, and to contribute with more data on the emission behaviour of biomass-burning boilers.

Particle emissions from a pellet stove, two pellet burners and two smaller biomass-fired district heating boilers were studied. Transient effects and influence of operating parameters, such as boiler load and excess air, were investigated. The district heating boilers were fired with wood pellets and wood briquettes and were equipped with multi-cyclones for dust removal. The particle emissions from the district heating boilers were measured downstream of the multi-cyclones. The particle emissions from the domestic heating devices were sampled in the chimney. Number concentrations and number size distributions were determined using an ELPI (Electrical Low-Pressure Impactor). In a few cases an ELPI was operated simultaneously with a SMPS (Scanning Mobility Particle Sizer) to analyse number concentrations and size distributions. Mass concentrations were measured by weighing of collected dust on filters according to Swedish standard. Mass size distributions were determined by a DLPI (Dekati Low-Pressure Impactor). In a few cases analysis of particle composition was carried out with focus on inorganic components. EDX (Electron Dispersive X-ray) analysis identified the main components. A new method to analyse the mass and molar fractions of alkali compounds, based on TOF-SIMS (Time-of-flight Secondary Ion Mass Spectrometry), was developed. CO, CO₂, O₂, THC (total hydrocarbons) were recorded in all cases, to give information about the combustion conditions. In most cases the NO_x-emission was recorded as well.

Mass concentrations of particles varied from 65 mg/m³ dry flue gas from the pellet stove to 84 mg/m³ dry flue gas for the district heating boiler fired with pellets, both emissions normalised to 10 % O₂. Number concentrations (30 nm - 10 µm) were in the range 10⁷ – 10⁸ cm⁻³. The particle emissions were dominated by submicron particles (particle diameter < 1 µm), both from number and mass perspective. The EDX analysis showed that the main inorganic components of the sub-micron particles were potassium, sulphur, chlorine and oxygen. Smaller amounts of sodium, magnesium and zinc were also found. Potassium sulphate was the most common compound and the second most common was potassium chloride.

Keywords: particle emissions, biomass combustion, domestic heating, district heating

PREFACE

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CONTENT

INTRODUCTION	1
Background	1
Literature Survey - Particles from Biomass Combustion	1
Particles from Incomplete Combustion	1
Ash Particles	3
Previous Measurements	4
Formation Mechanisms for Ash Particles during Biomass Combustion	18
Summary of the Literature Survey	21
The Objective of this Study	21
EXPERIMENTAL	22
Combustion devices	22
Grate Fired Boilers.....	22
Domestic Pellet Burners	23
Pellet Stove	23
Fuels	24
Measurement Set-Up	26
Particle Measurements	27
Gas measurements	30
Measurement Plan	31
RESULTS	34
Emission Characteristics of the Different Cases	34
The Influence of Operating conditions and Fuel Quality	38
Analyses of the Particle Composition.....	40
DISCUSSION	43
Mass, Number Concentrations, and Size Distributions	43
The Influence of Operating Conditions and Fuel Quality	44
Analyses of The Particle Composition	47
CONCLUSIONS	48
REFERENCES	49

Appendix 1 DESCRIPTION OF THE TOF-SIMS ANALYSIS

Appendix 2 SEPARATION CHARACTERISTICS OF MULTICYCLONES

INTRODUCTION

A short background stating the significance of the present study is given below followed by an overview of particle emissions from combustion of solid fuel. Finally, previous work on characterisation of particle emissions from biomass combustion is presented in this introduction.

Background

Biomass combustion for heat and power production is an important renewable energy source in an ecologically sustainable society. However, combustion involves emissions of particles and other air pollutants. In Sweden, combustion of biomass is increasing and this may lead to locally increased particle concentrations in the ambient air. Numerous epidemiological studies have shown correlations between negative health effects and increased concentrations of particulate matter in the ambient air (Dockery *et al.*, 1993, Pope *et al.*, 1995). Because of these negative health effects and the simultaneous increase of biomass combustion, the nature of particle emissions from biomass combustion needs further investigation.

The knowledge of the specific air pollution effects of particles from biomass combustion also needs to be improved. Particle concentrations are often reported as mass concentrations. However, Lighty *et al.* (2000) recently pointed out that particle surface area, number of ultrafine particles, bioavailable transition metals, polycyclic aromatic hydrocarbons (PAH), and other particle-bound organic compounds probably are more important than particle mass for the health effects of combustion particles. To improve the understanding of the air pollution effects related to particles from biomass combustion, there is a need to characterise these particle emissions in more detail.

Literature Survey - Particles from Biomass Combustion

This work is limited to primary combustion particles. Primary particles are emitted directly to the atmosphere from the combustion device (Smith and Sloss, 1998). There are two main sources of primary combustion particles from small-scale biomass combustion:

1. Particles from incomplete combustion. This includes soot, condensable organic particles (tar), and char particles.
2. Particles from the inorganic material in the fuel – ash particles.

In addition to primary particles, secondary particles, formed in the flue gas plume or in the atmosphere contribute to particle emissions. Secondary particles from biomass combustion can be made up of nitrates from nitrogen oxides, sulphates from sulphur dioxide and condensable organic particles. Thus condensable organic particles can be found both in primary and secondary combustion particles.

Particles from Incomplete Combustion

Particles from incomplete combustion represent not only hazardous emissions but also a fuel loss. Incomplete combustion occurs when the conditions allow gas-phase condensation reactions of the fuel and its decomposition products to compete with decomposition and

oxidation. Emissions of gaseous hydrocarbon compounds, soot, condensable organic particles, and char particles are a result of incomplete combustion.

Soot consists chemically of solid carbon. The soot formation mechanisms are complex and the current understanding of formation is incomplete. In Figure 1, a schematic description of soot formation is shown (for further details see e.g. Bockhorn (1994)). Soot formation starts during devolatilisation and combustion of volatiles when hydrocarbon fragments leave the fuel particles. These fragments then crack into smaller pieces and react with one another and surrounding gases. Aromatic rings are formed from these hydrocarbons, and these ring structures are thought to add alkyl groups, developing into polynuclear aromatic hydrocarbons (PAH). Thereafter, the particles grow larger by surface growth and agglomeration. The resulting soot particles are thus composed of agglomerates of smaller spherical particles, as indicated in Figure 1. This soot structure has also been observed experimentally (e.g. Bockhorn, 1994).

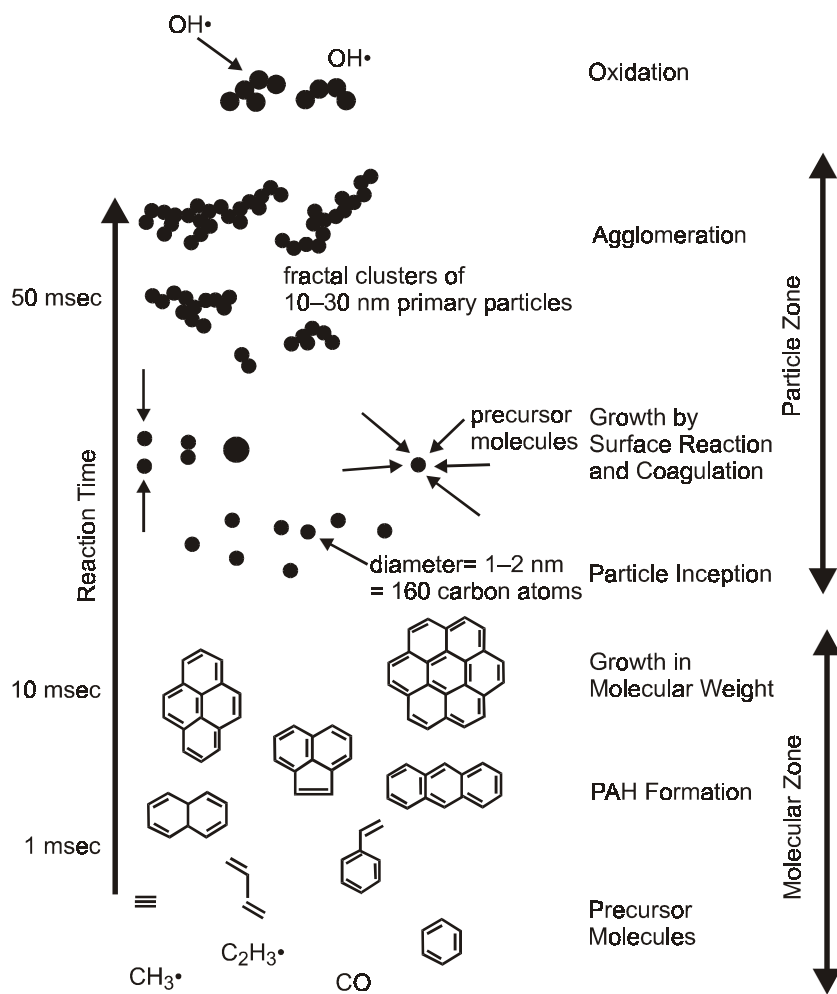


Figure 1 Schematic picture of soot formation (Bockhorn, 1994).

At low combustion temperatures and insufficient air supply (such as in certain old wood boilers), the emissions of tars can be significant. Condensable organic particles are found if the temperature has been low enough for the tars to condense. Tars are partly emitted as gases, which condense to particles in the plume. If the tars are included or not in a measurement depends on the measurement strategy. Often, particle sampling is made at higher temperatures or at temperatures where no condensation or only a part of the

compounds may have condensed, thus excluding condensable organic particles from the results. To include the tars in the particle measurements it is necessary to use a dilution tunnel or a similar device. Then, the sum of primary and secondary particles is measured. Otherwise, only primary particles are measured, since primary particles are directly emitted to the atmosphere from the source (Smith and Sloss, 1998). Char particles, on the other hand, consist of devolatilised fuel fragments.

Ash Particles

Ash emissions originate from the inorganic part of the fuel, and always remain as a by-product of combustion, regardless of complete or incomplete combustion. The ash-forming mechanisms of coal combustion are well established (Quann and Sarofim, 1982, Neville and Sarofim, 1982, Flagan and Friedlander, 1978), but the corresponding mechanisms in biomass combustion need to be further investigated.

Coal and biomass are two rather different solid fuels. While biomass recently was a living plant, it has taken several millennia for coal to be formed. The process of coal formation is slow and complex, involving increasing pressure and heat with time. Our present coal reserves were formed from vast peat swamps (Bryers, 1996). This difference results in that biomass in general has higher oxygen content and less ash, fixed carbon, sulphur and nitrogen than coal. The volatile fraction in biomass is high (over 80 % on an ash and moisture free basis). Furthermore, biomass ash reflects the trace minerals required for plant growth, while the coal ash tends to be composed of mineralogical material. For coal combustion, the ash forming species can be divided into three groups according to Sarofim and Helble (1993):

I	Included minerals	Mineral inclusions (formed at the same time as the coal, for example kaolinite $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot \text{H}_2\text{O}$);
II	Excluded minerals	E.g. shale or calcite pieces in the coal (or sand or clay from harvesting the biomass);
III	Inherent minerals	Organically bound or atomically dispersed inorganic material (components dispersed in the organic matrix).

In addition to this classification, ash-forming components can also be classified as easily leachable salts (Zevenhoven- Onderwater, 2001). The bulk of wood ash is composed of calcium, and the potassium content is typically 5 - 10 % (Bryers, 1996). Calcium has an important function in stiffening the cell walls of a plant. Potassium occurs as K^+ and is highly mobile with little structural function.

Despite the differences, both coal and biofuel are solid fuels and share some ash-transformation mechanisms (Baxter, 1993; Bryers, 1996). Ash particles from solid fuels are formed in two basic ways (Figure 2). The submicron particles are formed through condensation or heterogeneous or homogenous nucleation. Larger particles are formed through fragmentation during char burnout.

Formation of submicron ash particles starts with vaporisation, thermal or chemical disintegration of the inorganic material, convection during rapid devolatilisation or other reactions with organic material (Baxter, 1993), which result in inorganic vapours. From these vapours particles are formed by nucleation or condensation and growth by coagulation.

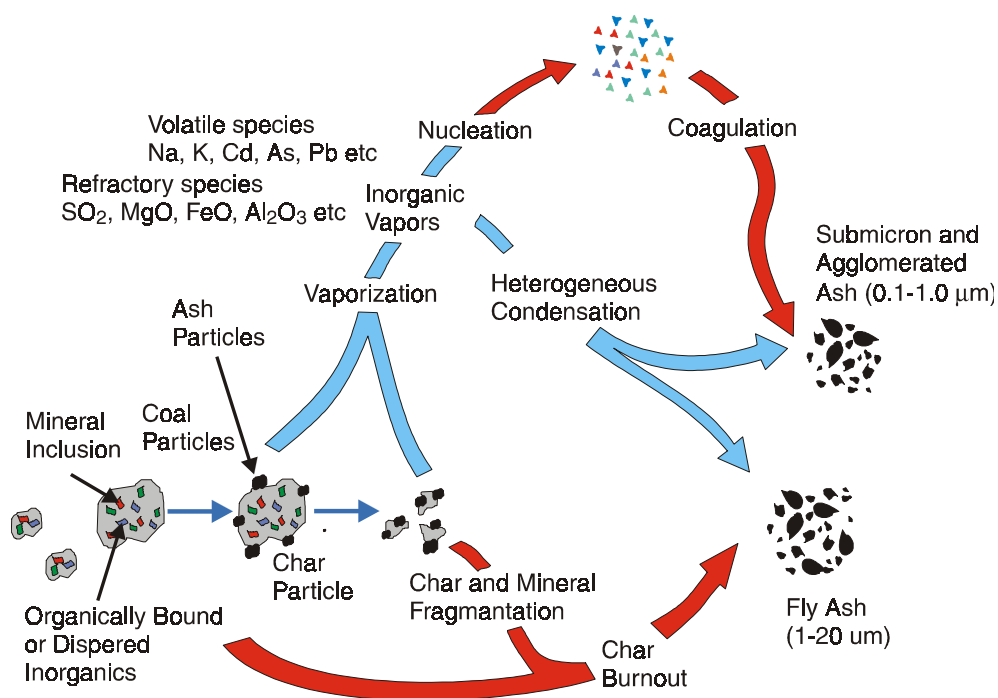


Figure 2 Mechanisms of formation of ash particles (Sarofim and Helble, 1993).

Larger ash particles are formed during char burnout. The residual ash then undergoes fragmentation either as a mineral grain or in conjunction with fragmentation of a burning char particle. It may coalesce with some or all of the remaining inorganic material, and it may undergo significant chemical or physical transformations. Release of atomically dispersed inorganic material from the fuel particle is affected by both the material's volatility and its reactions with the organic components in the fuel. Inorganic material that is inherently volatile at combustion temperatures includes derivatives of some of the alkali and alkaline earth metals, especially potassium and sodium. Non-volatile inorganic material can also be released from the fuel particle. Gases and tars then convectively carry the non-volatile material out of the fuel (Baxter *et al.*, 1992, cited by Baxter, 1993). Inorganic material in solid fuels may also come from extraneous sources. In coal this extraneous material is added to the fuel by the geologic processes and mining techniques. In biofuels, most of this material is added to the fuel during fuel processing in the field (Baxter, 1993). The extraneous material is often found as particles in nature and is the major contributor to fly ash particles larger than 10 μm . Sand, clay, and sea salt are examples of extraneous material in biomass.

Previous Measurements

Particle emissions from combustion are affected not only by the type of fuel and fuel quality but also of the boiler design, of the combustion conditions, and of the location in the combustion device where the measurements are performed. Below, results from previous measurements of particle emissions from biomass combustion are presented, starting with domestic heating devices, and ending with examples from large-scale biomass combustion.

Domestic Combustion of Biomass

Emissions from domestic combustion depend on combustion conditions. Favourable conditions mean sufficient temperature, air supply, residence time, and mixing. Unsatisfactory combustion conditions allow the gas-phase condensation reactions and the decomposition products of the fuel to compete with further decomposition and oxidation. In the latter case

the concentration of particles emitted may become high, and the emissions partly consist of soot, char, and organic condensable particles as a result of incomplete combustion. On the other hand, favourable combustion conditions lead to lower particle emissions because of no or minor emissions of particles from incomplete combustion. Inorganic compounds will then dominate the particle emissions.

Particle emissions from domestic combustion of wood pellets, wood logs and wood chips have been reported to be in the range 30 – 1 500 mg/m_n³ (Table 1). The highest mass concentrations were recorded (Muhlbaler Dasch, 1982; McDonald *et al.*, 2000) during combustion of wood logs in a wood stove or a fireplace. Muhlbaler Dasch (1982) investigated the emissions from different wood species and the influence of the size of the wood logs on the emissions. He observed that large wood logs led to higher particle emissions than split wood and showed that the higher particle emissions was almost entirely due to organic material. The combustion of large wood logs was related to unsatisfactory combustion conditions with low burning-rate, temperature and gas flow rates. Rau (1989) showed that such conditions were related to particle emissions dominated by organic material. He showed that for sub-stoichiometric conditions and at low temperatures the particle emissions from a wood stove mainly consisted of organic carbon. However, at higher temperatures, the aerosol composition contained less carbon (primarily elemental) and large amounts of potassium, chlorine and sulphur.

One has to be careful while interpreting literature data because the sampling method can affect the measured particle concentrations and size distributions. The sampling is carried out either in a flue gas channel or in a dilution tunnel. The purpose of a dilution tunnel is to imitate what happens to the flue gas when it reaches the ambient air, and the flue gas is diluted with large amounts of room-tempered air prior to sampling. This dilution and the residence time in the tunnel influence the particle concentration and the particle size distribution. The dilution cools the flue gas and causes formation of particles by homogenous or heterogeneous nucleation, condensation, and growth of particles by surface condensation. The residence time in the dilution tunnel affects the particle size distribution by facilitating particle growth by coagulation. Measurements by Wieser and Gaegauf (2000) both in a flue gas duct and a dilution tunnel illustrate this phenomenon, as can be seen in Table 1, for example, in the results from combustion in a pellet burner (third result from above compared to third result from below). Secondary formation and growth of particles is particularly important under unsatisfactory combustion conditions when high concentrations of organic compounds are emitted.

The largest mass concentrations in the tests mentioned above (Muhlbaler Dasch, 1982; McDonald *et al.*, 2000) were obtained under unsatisfactory combustion conditions. They were measured in the end of a dilution tunnel or in a similar way. Consequently, these emission levels represent concentrations in flue gas just cooled to ambient temperature and can be considered as examples of worst cases.

Total mass concentration is a common way to characterise particle emissions. The emitted particle mass may also be presented as mass size distribution (Table 2). The unsatisfactory combustion condition in the residential wood fireplaces investigated by Muhlbaler Dasch (1982) led to a maximum in the mass size distribution at a particle size of about 0.17 µm. This agrees with Oser *et al.* (2001) who reported a fine particle maximum somewhere less than 0.41 µm (i.e. maximum was below detection limit of instrument) during combustion of wood chips. However, Oser *et al.* (2001) found a bi-modal mass size distribution with a coarse

maximum at a particle size of about 10 μm . Combustion of small wood samples have shown a mode between 0.1 μm and 1 μm in the emitted wood smoke (Silva *et al.*, 1999).

Table 1 Mass concentrations of particles emitted from residential biomass combustion.

Residential heating				
Mass conc. (mg/m_n^3)	Fuel (ϕ= moisture)	Boiler type	Thermal output (kW)	Reference
33-120 ^a	Wood logs, beech, ϕ =16%	Room-heater	6 ^c	(Wieser and Gaegauf, 2000)
182-296 ^a	Wood logs, beech, ϕ =16%	Heat acc. soapstone stove	6 ^d	(Wieser and Gaegauf, 2000)
33-67 ^a	Wood pellets, ϕ =8%	Pellet burner	16	(Wieser and Gaegauf, 2000)
205-239 ^a	Wood chips, ϕ =60%	Wood chips burner (stoker)	70	(Wieser and Gaegauf, 2000)
PM _{0.4} = 92-107 ^a	Wood chips, ϕ =60%	Wood chips burner (stoker)	70	(Wieser and Gaegauf, 2000)
26-56 ^a	Wood chips	21 boilers, 7 boiler types	<50	(Launhardt <i>et al.</i> , 2000)
207 \pm 74 ^a	Wood logs	Room heaters	-	(Gaegauf <i>et al.</i> , 2001)
493 \pm 130 ^a	Wood logs	Acc. stoves	-	(Gaegauf <i>et al.</i> , 2001)
83 \pm 32 ^a	Wood logs	Wood boiler	-	(Gaegauf <i>et al.</i> , 2001)
59 \pm 3.5 ^a	Wood pellets	Pellet boiler	-	(Gaegauf <i>et al.</i> , 2001)
160 \pm 9 ^a	Wood pellets	Pellet room heaters	-	(Gaegauf <i>et al.</i> , 2001)
277 \pm 38 ^a	Wood chips, dry	Wood chips boilers	-	(Gaegauf <i>et al.</i> , 2001)
140 \pm 20 ^a	Wood chips, ϕ =30%	Wood chips boilers	-	(Gaegauf <i>et al.</i> , 2001)
100-120 ^a	Wood chips	Automatic under stoker furnace	120	(Oser <i>et al.</i> , 2001)
PM _{2.5} =700-1500 ^b	Wood (USA)	Wood stove & fire place	-	(McDonald <i>et al.</i> , 2000)
207-306 ^b	Wood logs, beech, ϕ = 16%	Heat acc. soapstone stove	6 ^d	(Wieser and Gaegauf, 2000)
PM _{0.4} =187-281 ^b	Wood logs, beech, ϕ =16%	Heat acc. soapstone stove	6 ^d	(Wieser and Gaegauf, 2000)
48-72 ^b	Wood pellets, ϕ = 8%	Pellet burner	16	(Wieser and Gaegauf, 2000)
PM _{0.4} =22-28 ^b	Wood pellets, ϕ = 8%	Pellet burner	16	(Wieser and Gaegauf, 2000)
2000 ^e	Wood logs (hardwood, softwood and synthetic)	Residential fireplaces, field and laboratory	-	(Muhlbaler Dasch, 1982)

^a The measurement point was located in the flue gas duct. In some cases the flue gas was diluted before measurement.

^b The measurement point was located at the end of a dilution tunnel.

^c 3 kg fuel/ batch

^d 7.5kg fuel/ batch

^e Residential: an aluminium chimney extension was placed on the chimney and the sample probe was inserted through the port. Lab: The sampling port was located 2 m above the grate.

The above reported mass characteristics of particles emitted from wood combustion are usually mean values sampled during combustion of a few batches of wood logs. However, during combustion of wood logs, the emissions vary considerably, not only between different kinds of combustion devices, but also between different combustion phases. In batch wise

feeding, three stages can be identified (for example, Axell *et al.*, 1997) when burning wood logs, i.e.

1. Ignition and start-up;
2. The intermediate phase with stable conditions;
3. Burn-out and extinction.

The ignition and start-up phase is characterised by high emissions of unburnt (CO and hydrocarbons). During the intermediate phase, the combustion is relatively stable (depending on the design and operation of the stove or boiler) and the emission of unburnt is low. When all volatiles have been burnt off and the residual char burns, the CO concentration generally increases due to higher excess air ratio, which cools the combustion chamber. The number concentration of emitted particles changes during the progress of combustion. The number of emitted particles per cubic metre was found to be highest during the start-up phase, lower at the intermediate phase and lowest during burn-out (Hueglin *et al.*, 1997; Wieser and Gaegauf, 2000; Gaegauf *et al.*, 2001). The emitted number concentrations from domestic combustion show large variations and were reported to be in the range 10^7 - 10^{10} particles/cm³ (Table 3).

Table 2 Characteristics of mass size distributions of particles emitted from residential biomass combustion.

Residential heating				
Maximum of the size distribution (µm)	Fuel	Boiler type	Thermal output (kW)	Reference
Fine peak <0.41 Coarse peak= ~10 ^a	Wood chips	Automatic under stoker furnace	120	(Oser <i>et al.</i> , 2001)
Mass median diameter ~0.17 ^b	Wood logs (hardwood, softwood and synthetic)	Residential fireplaces, field and laboratory	-	(Muhlbaler Dasch, 1982)
Lab furnaces or smaller lab scale				
Maximum of the size distribution (µm)	Fuel	Boiler type	Thermal output (kW)	Reference
0.1 < D _{p(max)} < 1 ^c	Chaparral, red prome, cork tree, Chinese elm tree, tumbleweed	Burning was initiated with a Bunsen burner flame	-	(Silva <i>et al.</i> , 1999)

^a The measurement point was located in the flue gas duct. In some cases the flue gas was diluted before measurement.

^b Residential: an aluminium chimney extension was placed on the chimney and the sampling probe was inserted through the port. Lab: The sampling port was located 2 m above the grate.

^c The smoke emitted was collected in a glass sampling bottle and analysed by aerosol time-of-flight mass spectrometry (ATOFMS).

Number size distributions strongly depend on the stage of wood combustion in a stove (Figure 3). The maximum of the number size distribution is found at the largest particle size during start-up. The maximum is displaced towards smaller particle size during the intermediate phase, and finally towards even smaller particle size during the burn-out phase (Hueglin *et al.*,

1997; Wieser and Gaegauf, 2000; Gaegauf *et al.*, 2001). Number particle size distributions in the flue gas from domestic wood boilers have been reported to be approximately constant and peak in concentration around a particle size of 70-90 nm (Schmatloch *et al.*, 2000; Gaegauf *et al.*, 2001). This is close to results from wood pellet boilers and wood chip boilers, yielding a number size distribution peaked at around 80 nm (Gaegauf *et al.*, 2001). It has been found that flue gases from residential wood boilers are dominated by submicron particles (Schmatloch *et al.*, 2000; Watson and Chow, 1999). Probably, these wood boiler measurements were carried out during the intermediate combustion phase of wood, which is considerably longer than the start-up and burn-out phases. A batch of wood logs in a stove is smaller than a wood batch in a boiler and also burns longer. Consequently, the different combustion phases of wood combustion do not affect the mean particle size distribution from wood combustion in a boiler as much as in a stove.

Table 3 Number concentrations of particles emitted from residential biomass combustion.

Number concentration (particles/cm ³)	Fuel (ϕ = moisture)	Boiler type	Thermal output (kW)	Reference
PM=1.06*10 ¹⁰ -1.67 *10 ¹⁰ a	Wood logs, beech, ϕ =16%	Room-heater	6 ^d	(Wieser and Gaegauf, 2000)
Start-up phase=4.4*10 ⁷ Intermed. phase=7.8*10 ⁷ Burn-out phase= 1.5*10 ⁷ a	Wood logs ϕ =15-18%	Wood stove	-	(Hueglin <i>et al.</i> , 1997)
3.11*10 ⁷ (O ₂ =6.2%), 10.1*10 ⁷ (O ₂ =11.6%) a	Wood chips, ϕ =45-70%	Wood chip burner	-	(Hueglin <i>et al.</i> , 1997)
“Vortex burner”: 2.17*10 ⁹ (O ₂ =7.0%) “Commercial boiler”: 3.57*10 ⁹ (O ₂ =7.9%) a	Wood chips	“Vortex burner” “Commercial boiler”	8.7	(Gaegauf and Wieser, 1998)
PM _{0.6} = 1.6 10 ⁸ a	Wood logs	Room heaters	-	(Gaegauf <i>et al.</i> , 2001)
PM _{0.6} =4.54 10 ⁸ a	Wood logs	Acc. stoves	-	(Gaegauf <i>et al.</i> , 2001)
PM _{0.6} = 1.8 10 ⁸ a	Wood logs	Wood boiler	-	(Gaegauf <i>et al.</i> , 2001)
PM _{0.6} = 0.8 10 ⁷ a	Wood pellets	Pellet boiler	-	(Gaegauf <i>et al.</i> , 2001)
PM _{0.6} = 2.5 10 ⁷ a	Wood pellets	Pellet room heaters	-	(Gaegauf <i>et al.</i> , 2001)
PM _{0.6} = 2.7*10 ⁸ a	Wood chips, dry	Wood chips boilers	-	(Gaegauf <i>et al.</i> , 2001)
Wood chips boilers PM _{0.6} =1.4*10 ⁸ a	Wood chips, ϕ =30%	Wood chips boilers	-	(Gaegauf <i>et al.</i> , 2001)
1*10 ⁸ -1*10 ⁸ (Increases when excess air ratios increases) a	Wood chips	Automatic under stoker furnace	120	(Oser <i>et al.</i> , 2001)
PM _{0.4} =0.765*10 ¹⁰ -6.58 *10 ¹⁰ b	Wood logs of beech, ϕ =16%	Room-heater (wood-fired)	6 ^c	(Wieser and Gaegauf, 2000)
PM _{0.4} =0.565*10 ¹⁰ -2.13 *10 ¹⁰ b	Wood logs of beech, ϕ =16%	Heat acc. soapstone stove	6 ^d	(Wieser and Gaegauf, 2000)
PM _{0.4} =23.8*10 ⁸ -28.,2*10 ⁸ b	Wood pellets, ϕ =8%	Pellet burner	16	(Wieser and Gaegauf, 2000)

^a The measurement point was located in the flue gas duct. In some cases the flue gas was diluted before measurement.

^b The measurement point was located at the end of a dilution tunnel.

^c 3 kg fuel/ batch

^d 7.5kg fuel/ batch

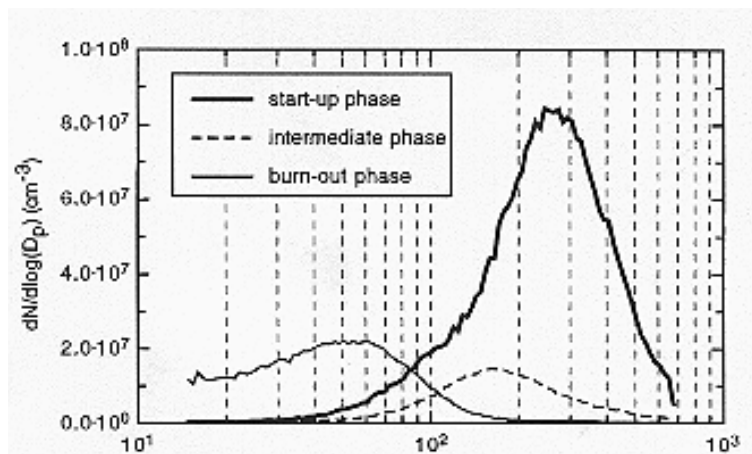


Figure 3 Number size distributions (size in nm) during the different phases in small-scale wood combustion. Reprinted with permission from (Hueglin *et al.*, 1997). Copyright (1997) American Chemical Society.

A wood boiler or a stove is manually fed batch-wise, whereas a wood chip burner is operated automatically. Wood chips are fed to a wood chip burner for instance by means of a screw. Nussbaumer and Hasler (1999) have stated that well designed and automatically operated wood furnaces may have a carbon concentration in the fly ash below 1 to 10 %. They also observed that the mass size distributions were unimodal, except for bark having a second maximum around 5 μm . Hueglin *et al.* (1997) observed two particle classes in the flue gas from a domestic wood chip burner. The dominant group was submicron particles with compact structures, but low amounts of semivolatile compounds were also present, typically considered as organic carbon. The second group was either perfectly spherical or consisted of irregularly shaped ensembles built of crystalline platelets.

Hueglin *et al.* (1997) considered the air supply to be an important parameter that affects the particle size distribution and the emissions of particle-bound polycyclic aromatic hydrocarbons (PAH). The oxygen concentration in the emitted flue gas was reported for a few cases of combustion in wood chip burners (Hueglin *et al.*, 1997, Gaegauf and Wieser, 1998, Wieser and Gaegauf, 2000, Gaegauf *et al.*, 2001, Oser *et al.*, 2001) and was found to affect the emissions, but it seems not to influence the number concentration or the maximum in the number size distribution of the emitted particles (Figure 4 and Figure 5). The maximum in number concentration, on the other hand, are probably affected by oxygen concentration because oxygen concentration is associated with favourable or unsatisfactory combustion conditions. In Figure 3, this is indicated by different maxima during the different combustion phases; largest particle size during lowest oxygen concentration, at start-up. However, these results (Hueglin *et al.*, 1997; Wieser and Gaegauf, 2000; Gaegauf *et al.*, 2001) lack information about oxygen concentration during the different combustion phases and are consequently not included in Figure 5. Note that there are not enough input data on mass concentration to investigate any trends associated with oxygen concentration in the publications quoted. To confirm trends between particle emissions and fuel quality or operation parameters more input data are needed.

Table 4 Maxima of number size distributions of particles emitted from residential biomass combustion.

Maximum of the size distribution (μm)	Fuel (ϕ = moisture)	Boiler type	Thermal output (kW)	Reference
Start-up phase= ~0.25 Intermediate phase= ~0.16 Burn-out phase= ~0.05 ^a	Wood logs ϕ =15-18%	Wood stove		(Hueglin <i>et al.</i> , 1997)
Geometric mean diameter=0.0686(O ₂ =6.2%) Geometric mean diameter=0.0957 (O ₂ =11.6%) ^a	Wood chips, ϕ =45-70%	Wood chip burner		(Hueglin <i>et al.</i> , 1997)
0.070 ^a	Wood logs, beech	Wood boiler	35	(Schmatloch <i>et al.</i> , 2000)
0.087 (2.7% O ₂), 0.080 (6.2% O ₂), 0.085 (7.9% O ₂), 0.060 (11.6% O ₂) ^b	Wood chips, ϕ =60%	Wood chip burner	-	(Wieser and Gaegauf, 2000)
0.1 ^a	Wood chips	“Vortex burner”	-	(Wieser and Gaegauf, 2000)
0.070-0.-0.090 ^a	Wood logs, ϕ =20%	Wood boiler	-	(Gaegauf <i>et al.</i> , 2001)
0.080 ^a	Wood pellets	Pellet boiler	-	(Gaegauf <i>et al.</i> , 2001)
Start-up phase= ~0.2 Intermediate phase= ~0.16 Burn-out phase= ~0.03 ^a	Wood logs	Wood stove	-	(Gaegauf <i>et al.</i> , 2001)
0.070 (6% O ₂), 0.072 (.93% O ₂), 0.065 (14%) ^a	Wood chips, ϕ =40%	Automatic under stoker furnace	120	(Oser <i>et al.</i> , 2001)

^a The measurement point was located in the flue gas duct. In some cases the flue gas was diluted before measurement.

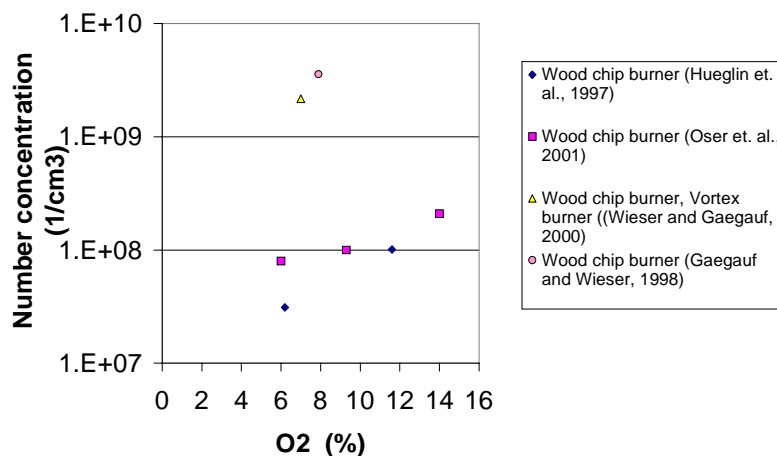


Figure 4 Oxygen concentration in the emitted flue gas plotted against number concentration of particles emitted from combustion in wood chip burners and measured in diluted flue gas from the stack.

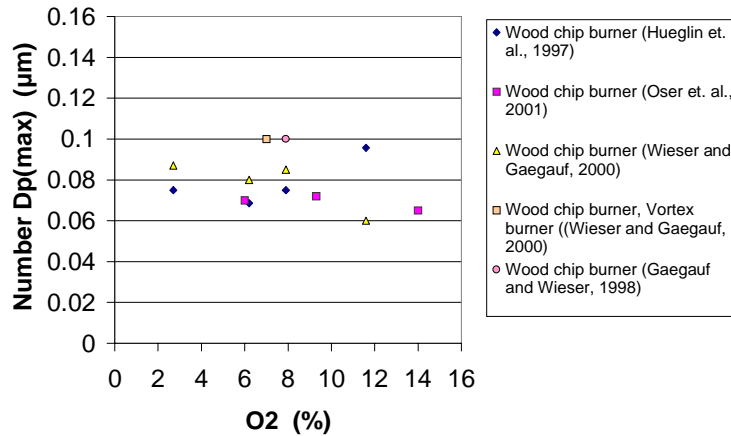


Figure 5 Oxygen concentration in the emitted flue gas plotted against maximum in the number size distribution of particles emitted from combustion in wood chip burners and measured in diluted flue gas from the stack.

The tables show available results from various sources. It is not straightforward to find trends, but despite the obvious statistical deficiencies some efforts to illustrate the data in diagrams will be made in the following figures. There is a slight tendency of a decreased number concentration of emitted particles for increasing moisture content in the fuel as shown in Figure 6. Results at different oxygen concentrations are plotted in series with constant moisture content in fuel. The influence of fuel moisture on the maximum in number size distribution is illustrated in Figure 7. However, no clear trends can be seen. The results from wood stove combustion differ, probably due to oxygen concentration. In this case the mass concentration appears to increase for increasing moisture content in the fuel (Figure 8). This can be an effect of lower combustion rate and unsatisfactory combustion due to increased moisture content in fuel.

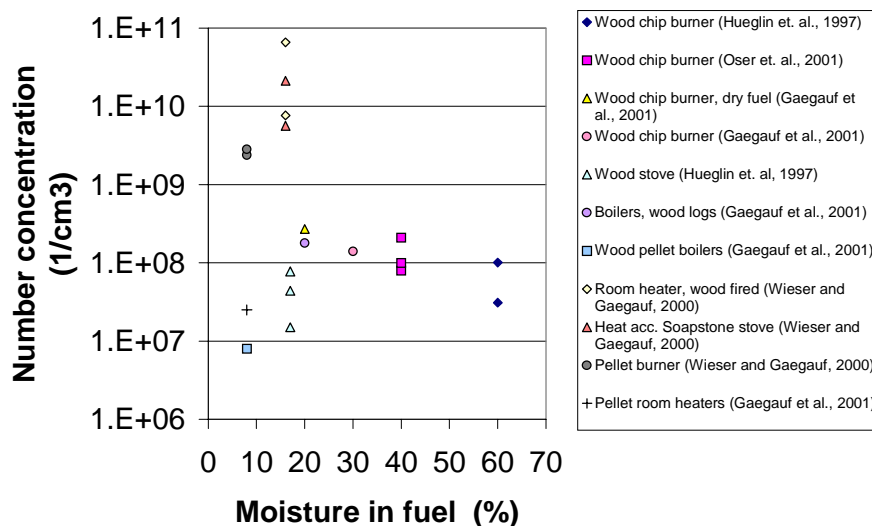


Figure 6 Moisture content in the fuel supplied plotted against number concentration of particles emitted.

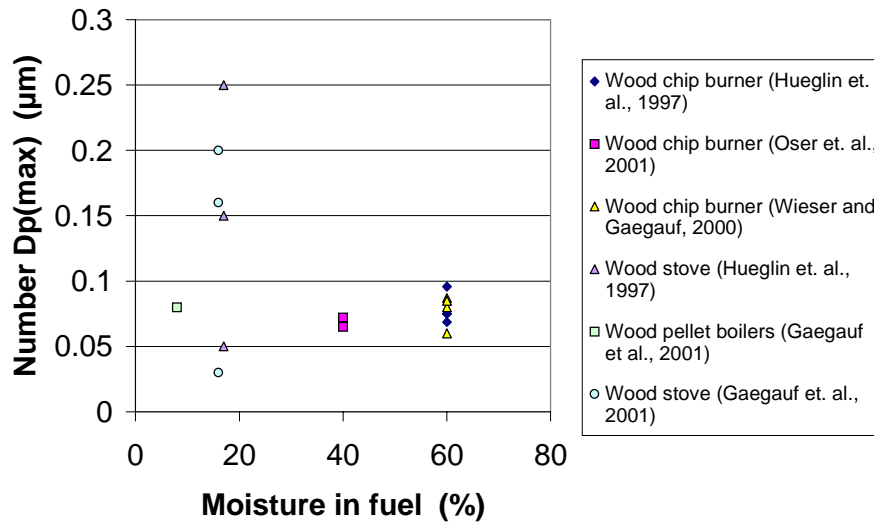


Figure 7 Moisture content in the fuel supplied plotted against maximum in number size distribution of particles emitted.

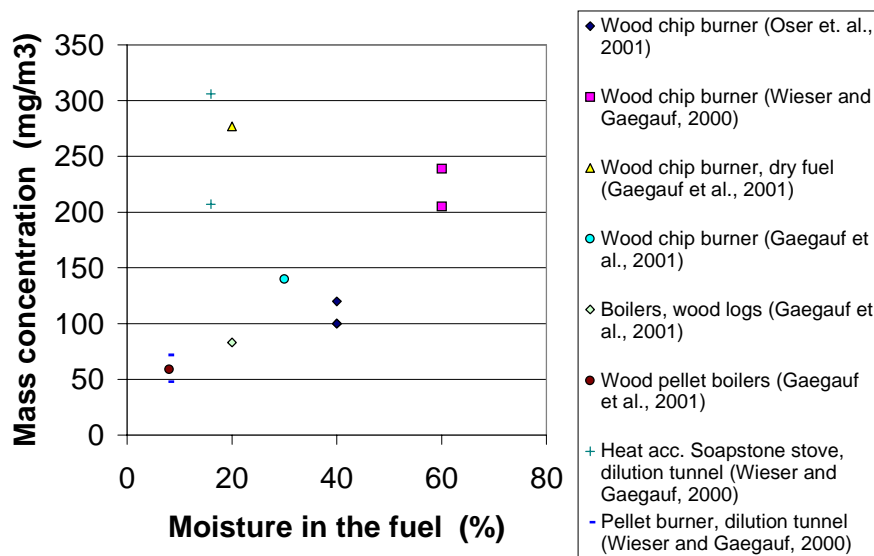


Figure 8 Moisture content in the fuel supplied plotted against mass concentration of particles emitted.

In summary: the mass and number concentrations in the flue gas from domestic biomass combustion stretch over a wide range. It seems that the large variations in particle emission characteristic are connected to variation in combustion conditions in the various combustion devices and during different combustion phases. Despite of the large variation in particle emissions from domestic combustion it is obvious that the emissions of submicron particles are dominant, both concerning mass and number concentrations. The peak of the number size distribution varies in a limited range. The maximum at smallest particle size of number size distributions occurred during burn-out in wood stoves and was reported to be around 0.03 µm. The maximum at largest particle size of number size distributions was found during start-up in wood stoves and was reported to be 0.2-0.3 µm.

Consequently, the combustion conditions are important for the number size distribution as well as for mass and number concentrations. Unsatisfactory combustion conditions sometimes arise because of poor fuel quality, for example, caused by high moisture content. The strong dependence on combustion conditions makes it important to use well operating combustion technique in order to avoid large mass concentrations of particles.

Large-Scale Combustion of Biomass

Normally, particle emissions from large-scale heat and heat and power plants are small because of favourable combustion conditions and dust-separation devices installed downstream of the combustion chamber. Soot emissions are typically negligible and emitted particles mainly consist of ash. The mass concentrations of particles, measured downstream of the convective pass (i.e. before particle separation), are reported to be in the range 60-3000 mg/m_n³ (Table 5). Several studies have shown bi-modal mass size distributions, where fine particles were only a minor part of the particle mass downstream of the convective pass (Table 5 and Table 6). However, there are also studies reporting uni-modal size distributions (Table 6).

The mass concentrations downstream of the convective pass from a circulating fluidised bed fired with willow (*salix*) and forest residue, were 200-400 mg/m_n³ (Lind *et al.*, 1997, Valmari *et al.*, 1998, Lind *et al.*, 1999). Measurements upstream, in, and downstream of a convective pass in the circulating fluidised bed boiler showed that a large mass share of supermicron particles was deposited in the convective pass (Lind *et al.*, 1997; Valmari *et al.*, 1998; Valmari *et al.*, 1999a; Valmari *et al.*, 1999b; and Lind *et al.*, 1999). Mass size distributions were bi-modal both upstream and downstream of the convective pass, and the size of the fine mode particles was in the range 0.1-0.6 µm and that of the coarse mode 1-100 µm. Coarse particles, i.e. particle diameters larger than one micron, were composed of irregular agglomerates. Fine mode particles were roughly spherical, and some of the largest fine mode particles were partially fused agglomerates of a few fine particles. A few ultrafine, rectangularly shaped particles were also observed by Valmari (1998) and Lind (1999). Submicron particles sampled upstream of the convective pass (830 °C), during combustion of forest residue, were found to be composed of sulphates and chlorides of potassium with some calcium and sodium. The main compounds were alkali sulphates (Lind *et al.*, 1997). Downstream of the convective pass (160 °C) of the boiler, the particles less than 0.5 µm mainly consisted of potassium chloride (Lind *et al.*, 1999). The coarse particles formed from the non-volatilised ash residue contained the major chemical elements calcium, potassium, phosphorous, magnesium and sulphur. With regard to the distribution of trace elements, Lind *et al.* (1999) reported that the major fraction of heavy metals was found in the coarse fly ash particles upstream of the convective pass (830 °C). Lind *et al.* (1999) also did not observe any enrichment of heavy metals in the fine particles downstream of the convective pass.

Formation of submicron particles during straw combustion has been investigated by Christensen and Livbjerg (1996), Christensen (1995), and Christensen *et al.* (1998). Several field measurements were carried out in two commercial scale CHP (combined heat and power) plants upstream of the fly ash filter (120 °C). Chemical analyses showed small soot amounts, so small that they were considered insignificant. Consequently, the particle emissions were mainly composed of ash. The emitted particle concentrations varied between 75 and 2095 mg/m_n³, and it was concluded that the mass loading of submicron particles was strictly correlated with the potassium content (Christensen, 1995). Measurements of the

aerosol and stack gas composition showed that a minimum of 19 % of the potassium was devolatilised. The corresponding numbers for chlorine and sulphur were 66 % and 43 %, respectively. The particles were reported to consist of almost pure KCl and K₂SO₄ with minor amounts of sodium, phosphorous and calcium. The number geometric mean diameter¹ of the particles was 0.15-0.30 µm with a typical number concentration of 1-2·10⁷ particles/cm_n³ (Table 7).

Table 5 Mass concentrations of particles in biomass-fired district heating boilers

Large scale boilers				
Mass conc. (mg/m_n³)	Fuel	Boiler type	Thermal output (MW)	Reference
Total= 190-270 ^a	Swedish willow (salix)	CFB	35	(Lind <i>et al.</i> , 1997)
Total=230-320 ^a Fine (<0.5 µm)= 14-23 ^a	Swedish forest residue	CFB	35	(Lind <i>et al.</i> , 1999)
PM ₁ = 75-2095 ^a	Straw (12 types)	-	18 / 25	(Christensen, 1995)
Total=400 ^a Fine mode=50-90 ^a	80% willow (salix), 20% wood pellets	CFB	12	(Valmari <i>et al.</i> , 1998)
Total = 530-620 ^b	Swedish willow (salix)	CFB	35	(Lind <i>et al.</i> , 1997)
Total=1000-1300 ^b Fine (<0.5 µm)= 23-25 ^b	Swedish forest residue	CFB	35	(Lind <i>et al.</i> , 1999)
Coarse mode= ~600 ^b Fine mode= 50-80 ^b	80% willow (salix), 20% wood pellets	CFB	12	(Valmari <i>et al.</i> , 1998)
Smaller district heating plants				
Mass conc. (mg/m_n³)	Fuel	Boiler type	Thermal output (MW)	Reference
Wood chips=60-230 ^a Bark=450-480 ^a	Wood chips, bark	Moving grate	0.44	(Obenberger <i>et al.</i> , 2001)

^a The measurement point was located downstream of the convective path.

^b The measurement point was located at high temperature, upstream of or in the convective path.

Results, similar to those from straw combustion in heat and power plants, were reported by Kaufmann and Nussbaumer (1998) on combustion of herbage-grass (hay) and straw in smaller grate-fired furnaces (50, 100 and 450 kW_{th}). The particles were in general less than 0.2 µm with KCl as the dominant compound. The main elements in the fly ash from herbage-grass were potassium (30 %), chlorine (24 %) and sulphur (6 %). The amount of carbon was small, indicating efficient burn-out. The main components were inorganic and the particle emissions principally consisted of ash.

¹ The geometric mean, d_g , is defined (Hinds, 1999) as the Nth root of the product of N values:

$$d_g = (d_1 d_2 d_3 \dots d_N)^{1/N}$$

$$\text{For grouped data with I intervals: } d_g = (d_1^{n_1} d_2^{n_2} d_3^{n_3} \dots d_I^{n_I})^{1/N}$$

Table 6 Maxima of mass size distributions of particles emitted from district heating boilers.

Large scale boilers				
Maximum of the size distribution (μm)	Fuel	Boiler type	Thermal output (MW)	Reference
Fine=0.2-0.3, Coarse= 3 ^a	Swedish willow (salix)	CFB	35	(Lind <i>et al.</i> , 1997)
Fine d(mean)=0.2 Coarse d(mean)= 3 ^a	Swedish forest residue	CFB	35	(Lind <i>et al.</i> , 1999)
Mass Geometric Mean=0.4 ^a	Straw (12 types)	-	18 / 25	(Christensen, 1995)
Fine mode=0.1-0.6, Coarse mode=1-100 μm ^a	Swedish forest residue	CFB	35	(Valmari <i>et al.</i> , 1998)
Fine =0.2-0.3, Coarse= 6 ^b	Swedish willow (salix)	CFB	35	(Lind <i>et al.</i> , 1997)
Fine d(mean)=0.1, Coarse d(mean)=12 ^b	Swedish forest residue	CFB	35	(Lind <i>et al.</i> , 1999)
Fine mode=0.1-0.6, Coarse mode=1-100 μm ^b	Swedish forest residue	CFB	35	(Valmari <i>et al.</i> , 1998)
Smaller district heating plants				
Mode (μm)	Fuel	Boiler type	Thermal output (MW)	Reference
Max=0,14 ^a	Wood chips	Grate	1.4 / 3 / 2.5	(Machan, 1998)
Wood chips: Max ~0.1 Bark: Max ~ 0.2 ^a	Wood chips, bark	Moving grate	0.44	(Obenberger <i>et al.</i> , 2001)
Typical size < 0.2 ³	Herbage-grass (hay), straw, and wood	Under feed stoker / Grate	0.1 / 0.45	(Kaufmann and Nussbaumer, 1998)

^a The measurement point was located downstream of the convective path.

^b The measurement point was located at high temperature, upstream of or in the convective path.

³ The measurement point is **assumed** to be downstream of the convective path.

Table 7 Number concentration and maxima of number size distributions of particles emitted from district heating boilers.

Large scale/ smaller district heating				Reference
Number conc. (particles/cm_n³)	Fuel	Boiler type	Thermal output (MW)	
PM ₁ = 1-2 *10 ⁷ ^a	Straw (12 types)	-	18 / 25	(Christensen, 1995).
Maximum of the size distribution (μm)	Fuel	Boiler type	Thermal output (MW)	
Number geometric mean=0.15-0.30 ^a	Straw (12 types)	-	18 / 25	(Christensen, 1995).
Fine (<1) maximum = 0.1 ^a	Wood waste/Wood chips	CFB	23	Moisio (1999)
~0.09 ^b	Dry wood pellets	Grate	0.8-1.5	(Lillieblad <i>et al.</i> , 2000)
~0.10 ^b	Wet forest residue	Grate	0.8-1.5	(Lillieblad <i>et al.</i> , 2000)

^a The measurement point was located downstream of the convective path.

^b The measurement point was located downstream of the multicyclone.

In a detailed study of the chlorine emissions during combustion of herbs, Kaufmann (1997) has investigated the influence of oxygen concentration on particle emissions. At high excess air ratios ($O_2=11.4\%$) the typical particle size was $0.2-0.3\ \mu\text{m}$, whereas the typical particle size increased to $0.3-0.5\ \mu\text{m}$ at lower excess air ratios ($O_2=8.6\%$). Measurements of particle size mass distributions in smaller district heating plants during combustion of wood chips, sawdust, bark, fibreboard and waste wood (Machan et. al., 1998; Brunner *et al.*, 2000; Obenberger *et al.*, 2001), considering particles smaller than $10\ \mu\text{m}$, indicated unimodal size distributions with a maximum less than $1\ \mu\text{m}$ (Machan, 1998). The condensed phases consisted of KCl , K_2SO_4 , K_2CO_3 , NaCl and Na_2SO_4 .

Brunner *et al.* (2000) sampled particles during combustion of beech, bark, fibreboard and waste wood in two grate furnaces, $0.44\ \text{MW}$ and $3\ \text{MW}$, showing that the submicron particles mainly were composed of alkali salts in wood chips combustion, alkali salts and calcium in combustion of bark, and for waste wood there was a significant influence of heavy metals as zinc and lead. The coarse mode particles consisted of refractory species like calcium, silicon, magnesium and aluminium. The submicron particles appeared as spheres or crystals, while the coarse particles were amorphous (if molten) or consisted of a grid built up by refractory species on which smaller particles were agglomerated.

In two, 0.44 and $40\ \text{MW}_{\text{th}}$ grate furnaces, the particle emissions during combustion of chemically untreated wood chips, fibreboards, bark, and waste wood have been studied (Obenberger *et al.*, 2001). The measurements were performed at the boiler outlet, i.e. before dust separation and where the flue gas was $180-220\ ^\circ\text{C}$, and showed a correlation between high ash content in the fuel and a high particle concentration in the flue gas. Combustion of wood chips resulted in concentrations of fly ash in the range $60-230\ \text{mg}/\text{m}_n^3$ (in dry flue gas at $13\% \text{CO}_2$). For waste wood combustion the fly ash concentrations were about $300\ \text{mg}/\text{m}_n^3$, whereas for bark or fibreboard the concentrations were in the range $450-500\ \text{mg}/\text{m}_n^3$. The mass size distribution in the particle size range less than $10\ \mu\text{m}$ showed uni-modal mass size distributions. Wood chips combustion led to a peak in the mass size distribution at $0.1\ \mu\text{m}$. For bark combustion the peak was found at $0.2\ \mu\text{m}$ and for waste wood at $0.4\ \mu\text{m}$.

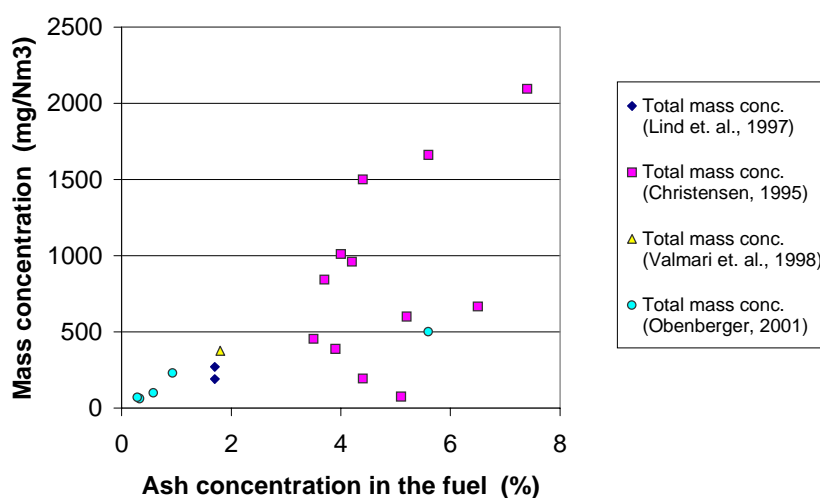


Figure 9 Ash concentration in the fuel plotted against mass concentration in flue gas downstream the convective pass.

A number of previous investigations have presented fuel analyses together with emission characteristics. The ash concentration in the fuel seems to correlate with mass concentration of particles in the flue gas downstream of the convective pass (Figure 9). It is also indicated that increased potassium, chlorine, and sulphur concentrations in the fuel lead to increased mass concentrations downstream the convective pass (Figure 10, Figure 11, and Figure 12). These trends consequently indicate that fuel composition may be important for the composition of particle emissions.

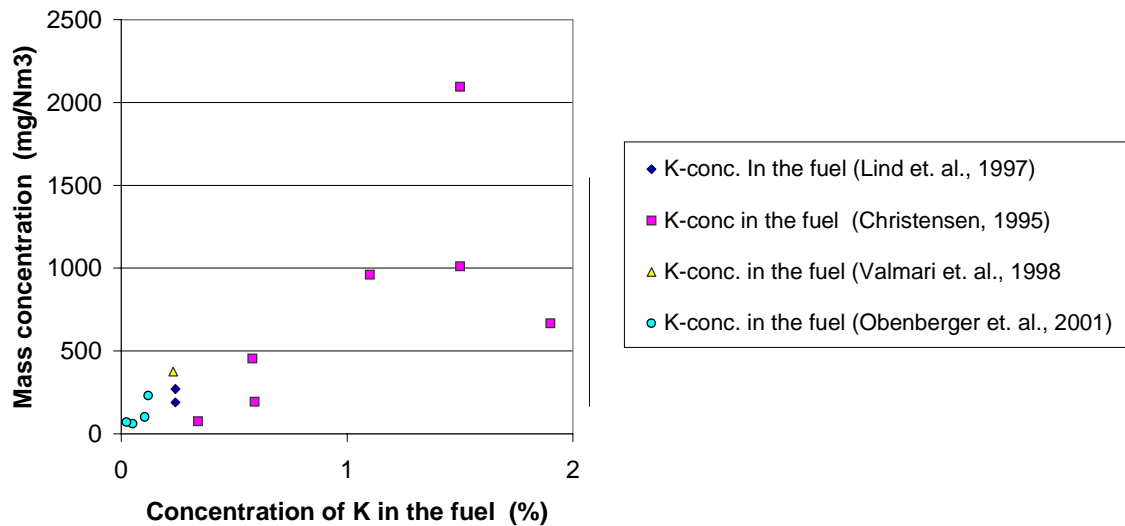


Figure 10 Potassium concentration in the fuel plotted against mass concentration in flue gas downstream the convective pass.

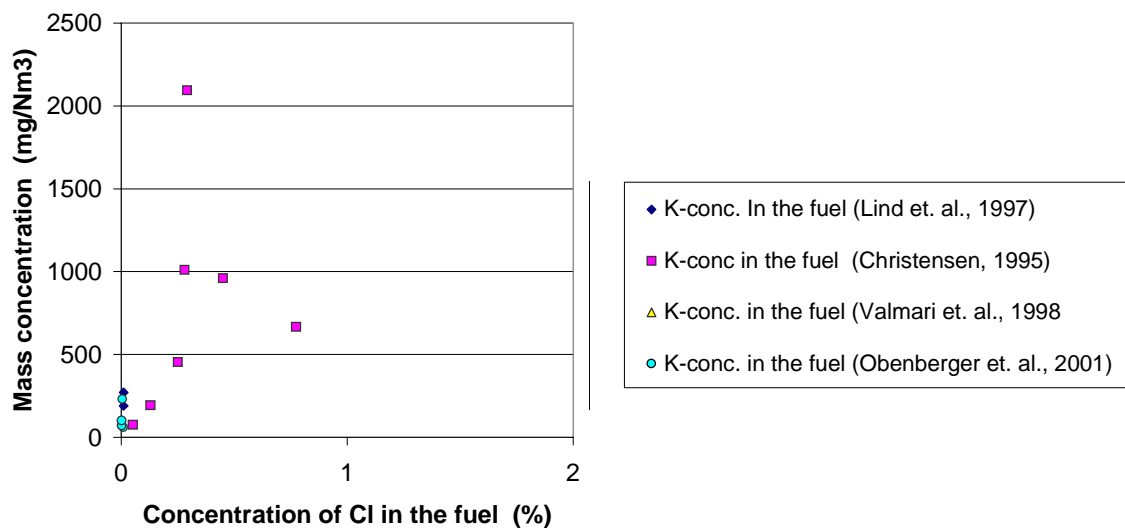


Figure 11 Chlorine concentration in the fuel plotted against mass concentration in flue gas downstream the convective pass.

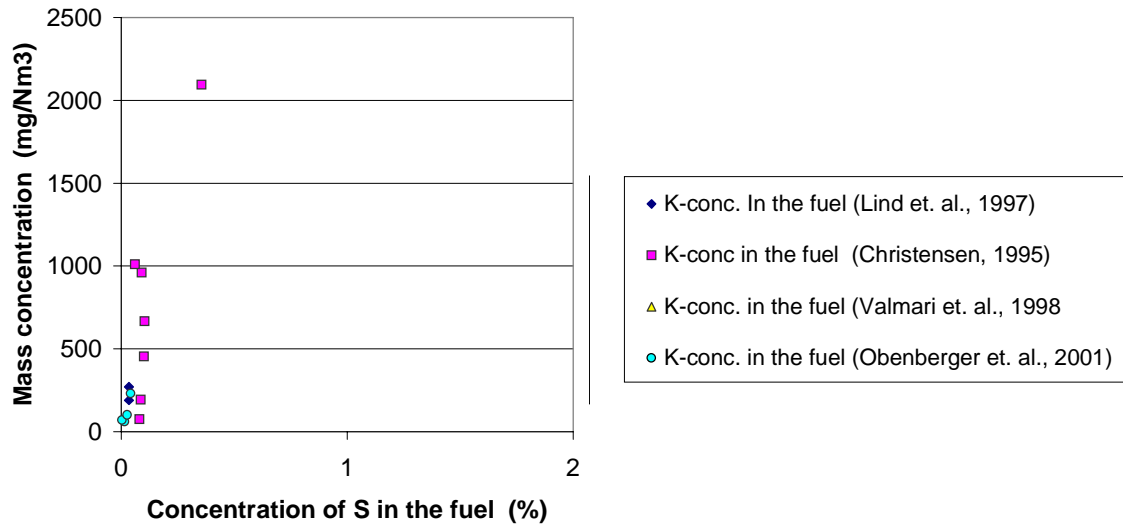


Figure 12 Sulphur concentration in the fuel plotted against mass concentration in flue gas downstream the convective pass.

In summary: for larger boilers, several particle studies downstream of the convective pass show a great variation of mass concentrations of emitted particles. The characteristics of the mass size distributions appear to be rather constant in the submicron range. However, both uni-modal and bi-modal mass size distributions have been reported in studies that involve both submicron and supermicron particles. All investigations point out potassium chloride as the main compound in the submicron range, but potassium sulphate is also important. Furthermore, minor amounts of sodium, phosphorus, calcium, zinc, and lead have been reported.

Formation Mechanisms for Ash Particles during Biomass Combustion

Previous field and laboratory studies of the particle emissions from biomass combustion point out potassium but also sodium as important components in ash particles. Thus, more basic knowledge about the behaviour of the alkali metals during combustion is important for the understanding of the particle formation mechanisms of biomass particles.

Laboratory studies of pyrolysis of wheat straw have shown that there are two characteristic temperature intervals where the alkali metals are released (Olsson *et al.*, 1997). A small fraction of the alkali metals left the fuel below 500 °C, whereas the major part was released at temperatures above 500 °C. At higher temperatures, the particle emissions were found to correlate with the chlorine content. In a study of chlorine emissions during combustion of herbs and wood (Kaufmann, 1997) it is concluded that fly ash mainly is formed from evaporation of inorganic compounds such as KCl. It was also observed that compounds in the fly ash undergo chemical transformations, for example the transformation of KCl to K₂SO₄ in the presence of SO₂. Furthermore, Kaufmann (1997) suggested that HCl primarily forms during the devolatilisation stage but also as a by-product of the formation of K₂SO₄ from KCl. The release of alkali vapour during combustion and gasification of switchgrass was studied in a laboratory furnace. The initial feedstock composition was the most important factor for the amount and species of alkali metals that were released (Dayton *et al.*, 1995). The switchgrass contained high amounts of both potassium and chlorine, and the dominant alkali species

released was concluded to be potassium chloride. Combustion temperature, oxygen concentration in emitted flue gas, and moisture content in the fuel seem to have a small influence on the alkali release. Variations in the combustion conditions (800/1100°C, 5/20 % O₂, 10 % O₂/20 % H₂O) affected the alkali release by a factor of 2 or less (Dayton *et al.*, 1995). A study of the release of alkali vapour during combustion/gasification of switchgrass indicated that the composition of the fuel is the most important factor for the amount and species of alkali metals released (Dayton and Milne 1996). For mechanisms of alkali release during combustion of biomass was identified:

1. For woody fuel, which got relatively low alkali as well as chlorine content, alkali is primary released through vaporisation or decomposition of alkali sulphate.
2. During combustion of fuel with high concentration of alkali as well as chlorine, for example herbaceous fuel, grass, and straw, alkali metal chlorides are the primary species released.
3. For fuels with high concentration of alkali and low chlorine content, most of the alkali leaves the fuel as alkali hydroxide.
4. Combustion of fuel with high alkali content coupled with high levels of fuel-bound nitrogen lead to alkali vapour released as alkali cyanide as dominating compound.

Another study (Dayton *et al.*, 1995) showed that adding steam to the combustion environment shifted the release of alkali from alkali chloride to alkali hydroxide. Thus, this study indicates influence of fuel moisture.

A few detailed explanations of formation of biomass particles have been presented. For combustion of straw, Christensen (1995) developed a numerical model to describe formation and evolution of a multi-component aerosol during cooling of a gas containing condensable vapours. The model includes gas phase reactions. Based on the model, Christensen discusses a number of possible mechanisms for formation of fine mode aerosol during straw combustion:

1. KCl is partially converted to K₂SO₄ in the gas phase, the sulphate nucleates homogeneously and both species condense heterogeneously on the new particles.
2. KCl nucleates and condenses heterogeneously. The conversion to sulphate proceeds primarily through a complex gas-solid reaction catalysed by SO₃.
3. Nucleation of K₂CO₃ or K₂SO₃ takes place prior to the condensation of KCl and K₂SO₄. The potassium condenses as K₂CO₃, K₂SO₃ and KCl. K₂SO₃ and KCl react to form K₂SO₄.
4. Particle formation can take place at high temperatures if refractory metals (for instance calcium, magnesium, iron and silica) are volatilised during combustion. KCl and possibly K₂SO₄ may condense on these particles at lower temperatures as in mechanisms 1 and 2.

Obenberger *et al.* (2001) recently identified three processes of aerosol formation related to three bio-fuels: chemically untreated wood chips, bark, and waste wood. The mechanisms were developed from field measurements and the characterisations of the combustion particles in grate furnaces and are presented below:

1. **Wood chips:** Submicron particles from combustion of wood chips mainly consisted of potassium, sulphur and chlorine. The concentration of potassium in the emitted particles increased with decreasing particle diameter. The calcium content increased with increasing particle diameter for particles larger than 0.8 μm . It was concluded that potassium sulphates and chlorides were formed by homogenous gas phase reactions. These compounds then nucleated homogeneously and/or condensed on existing surfaces. Calcium was a part of the coarse particle fraction, but it interacted with the submicron fraction by offering surfaces for condensation of metal vapours. The concentrations of the heavy metals lead, zinc, arsenic and cadmium increased with decreasing particle size. From this observation Obenberger *et al.* (2001) suggested that the most probable formation routes of heavy metal aerosol particles were from gaseous phase to the solid phase and that the heavy metals condensed after the primary particles were formed.
2. **Bark:** Submicron particles from combustion of bark consisted of potassium, sulphur and chlorine, but also of considerable amounts of calcium and heavy metals, such as zinc and lead. It was proposed that very small calcium particles exist in the flue gas leaving the fuel bed, because thermodynamic data of calcium showed that release of calcium to the gas phase was not possible. Consequently potassium chloride and potassium sulphate probably condensed on the surface of the calcium particles, as well as nucleated homogeneously. Supermicron particles were found to contain increasing amounts of calcium and magnesium, which was explained by entrainment from the fuel bed with the flue gas. The heavy metal concentrations were higher than for wood chips (in analogy with the fuel analyses), but the same particle formation routes as discussed above for wood chips were suggested.
3. **Waste wood:** The waste wood contained considerably higher amounts of zinc and lead, which was assumed to affect the ash forming mechanisms. Elemental zinc in a reducing atmosphere can evaporate at high temperatures. Obenberger *et al.* (2001) proposed that zinc is released to the gas phase in this way and it afterwards reacts with oxygen. The vapour pressure of the resulting ZnO was very high and thus nucleation of ZnO was assumed to occur. The ZnO particles formed were then supposed to agglomerate. This explained why zinc concentration followed the particle size distribution and did not increase with decreasing particle size as in the case of wood chips and bark. Due to the large surface area of the ZnO particles, the gaseous alkali compounds and lead condensed on the surfaces of ZnO particles. The ash forming mechanisms of waste wood differ from the ash forming mechanisms corresponding to combustion of wood chips and bark suggested by the same authors.

Since soot emissions from large boilers are small, studies of formation of particles from biomass combustion have focused on inorganic particles, probably because these also cause deposition problems. Various mechanisms for formation of submicron ash particles have been suggested, but potassium chloride and potassium sulphate are in focus. These alkali particles are formed by homogenous and/or heterogeneous nucleation. In some cases, refractory species, such as calcium, are suggested to act as cores for heterogeneous nucleation of potassium sulphate and chloride.

Summary of the Literature Survey

It can be concluded from the preceding survey of literature that mass concentrations of particles from biomass combustion stretch over a wide range, even among domestic heating devices and between larger boilers. In domestic heating, favourable combustion conditions are important in order to avoid emissions of unburnt material. On the other hand, unburnt material in particle emissions from large boilers are reported to be negligible. Particle emissions from domestic heating are dominated by submicron particles. However, in large boilers, mass size distribution upstream of dust separation devices typically is bi-modal. Assuming well operating dust separation devices, the size distributions of particles emitted will be uni-modal, only containing a submicron mode. Alkali compounds, primary potassium compounds, dominate the submicron particles from biomass combustion. For large boilers, it has been found that increased potassium, chlorine, and sulphur concentrations in the fuel lead to increased mass concentrations of particles upstream of dust separation devices. The concentration of minerals in the fuel seems to correlate with the mass concentrations of particles upstream of the dust separation devices. Below, a summary is presented showing the influence of excess air ratio and moisture content of fuel on the emissions (see also Figure 4- Figure 8):

- The mass concentration increases under unsatisfactory combustion conditions due to particles originating from incomplete combustion. For example, unsatisfactory combustion conditions can be a result of low combustion rate due to high moisture content in the fuel or low excess air ratio.
- There is a slight tendency to a decreased number concentration of emitted particles for increasing moisture content in the fuel.
- Excess air ratio does not seem to influence the number concentration.
- Number size distribution seems to be independent of fuel moisture.
- Excess air affects the number size distribution. For example, during wood combustion the number maximum is at largest particle size during start-up, at smaller size during the intermediate phase, and at smallest size during burn-out.

In conclusion, the particle formation mechanisms are not yet fully understood. Differences in chemical composition of the fuel probably affect the formation mechanisms and this should be further investigated. For instance, very few detailed studies on refined fuels, such as wood pellets and briquettes, have been reported. The influence of combustion conditions on the particle formation mechanisms should also be further studied.

The Objective of this Study

This study aims at chemical and physical characterisation of the particle emission from combustion of wood pellets and wood briquettes in commercial domestic combustion devices and smaller district heating plants. The flue gas aerosol is characterised also by the gaseous emissions, because an important part of the work is to relate particle emissions to gaseous emissions and, thus, to combustion conditions.

EXPERIMENTAL

Particle emissions from small-scale combustion devices will be investigated by field and laboratory measurements. In this experimental section, first the combustion devices, second the fuels, third the measurement set-up, and finally the measurement plan are presented.

Combustion devices

Three types of devices designed for combustion of wood pellets and one designed for combustion of wood briquettes will be investigated. The sizes range from domestic heating to small-scale district heating.

Grate Fired Boilers

Particle emissions from two smaller grate-fired boilers are considered, one fired with wood pellets and the other with wood briquettes. The basic boiler designs are of the same kind, but there are nevertheless a few differences, which are explained below.

The pellet district heating boiler investigated is located in Sandared (near Borås in Sweden), and has a thermal output of 1.75 MW. A schematic picture of the boiler is shown in Figure 13. The boiler has a fixed grate with moving scrapes and is equipped with a multicyclone as dust separator. The fuel is fed forward by the moving scrapes and the feeding can be adjusted to six fuel flows. The primary air is supplied from below the grate and divided into three air zones. The primary air supply is adjusted according to the fuel supplied. Secondary air is introduced downstream of the grate and the secondary flue gas fan is controlled by the O_2 -concentration in the flue gases. The Sandared boiler is operated at full load and with an excess air corresponding to 9 % O_2 in the flue gas during the major part of the firing season. Normally, the boiler is operated with a certain degree of flue gas recirculation.

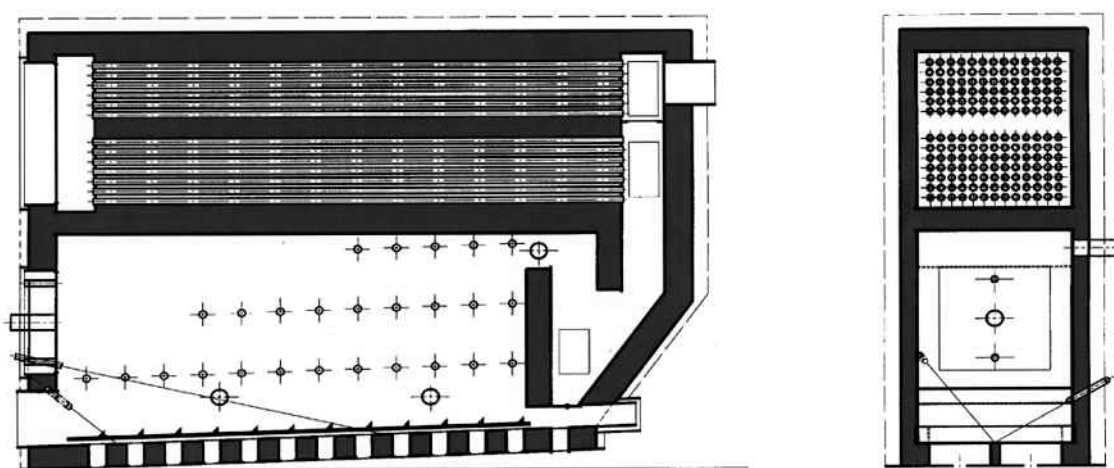


Figure 13 The Sandared boiler.

Another district heating boiler, located in Fristad (also near Borås in Sweden) was also studied. The Fristad boiler has a thermal output of 2.50 MW and is fired with wood briquettes. It is normally operated at full load with an excess air corresponding to 6 % O_2 in the flue gas.

The basic principles of the Fristad boiler are the same as those of the Sandared boiler. Though there are some minor differences, i.e. the Fristad boiler is designed without flue gas recirculation and with a tertiary air register besides primary and secondary air supply.

Domestic Pellet Burners

A domestic pellet burner is similar to an oil burner installed in a domestic boiler. The pellet burner is connected with a pellet storage. A schematic picture of a domestic pellet burner is presented in Figure 14. Pellets are transported upwards from the bottom of the pellet storage by a screw. Where the screw ends, the pellets fall through a tube to the burner cup. The combustion takes place in the burner cup.

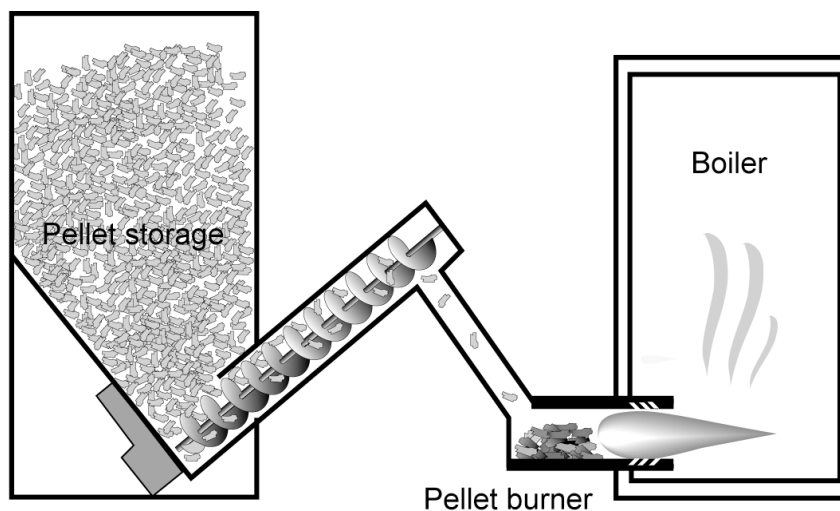


Figure 14 Schematic picture of a pellet burner

A pellet burner is installed in a boiler to heat a house. In the laboratory a cooling rig simulates the heat demand of a house. Normally, installed in a house, the boiler is set at thermostat control that results in a cyclic intermittent operation of the pellet burner. Depending on the design of the pellet burner the emission characteristics at intermittent operation differ. In this study one pellet burner with pilot flame (i.e. a small flame is maintained also when there is no heat demand) and one burner with electric ignition were considered. The first part of a cycle is similar in the two designs and is characterised by favourable combustion conditions. Eventually, the boiler thermostat switches off the burner. The second part of the cycle in a burner with pilot flame is characterised by a glow-bed with low flue gas flow and high concentrations of carbon monoxide and hydrocarbons. The second part of the cycle in a pellet burner with electrical ignition is characterised by no combustion at all. Beside the fact that one pellet burner was designed with pilot flame and the other one with electric ignition, there was one additional difference in burner design: The pellet burner with pilot flame burns with the flame upwards, and the burner with electric ignition burns with the flame forward.

Pellet Stove

The pellet stove of this study was equipped with an internal pellet storage (Figure 15). The pellets are transported upwards from the bottom of the storage by a screw. Then the pellets fall into a burner cup where combustion takes place. The flame is directed upwards and can be seen from the outside of the stove, since the front of the pellet stove has a glass window. The

flue gases are drawn out of the stove with a flue gas fan. The combustion air enters the combustion cup through a tube that ends up below the pellet store on the rear side.

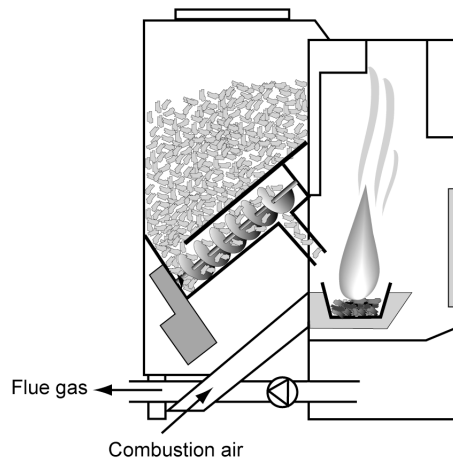


Figure 15 Schematic picture of the pellet stove.

Fuels

In this investigation, the focus is on refined fuels as pellets and briquettes. Pellets and briquettes are mainly produced from saw-dust and wood shavings. Consequently, these fuels are relatively homogeneous and have low moisture and ash contents. The properties of the fuels used in this study are presented in

Table 8 and Table 9. Wood pellets were fired as the main fuel in four of the combustion devices investigated. The wood pellets fired in the Sandared district heating boiler, in the pellet burners and in the pellet stove in the laboratory, had similar chemical composition. The chemical composition in the wood briquettes fired in the Fristad district heating boiler was also rather similar to that of the wood pellets. In Fristad, dried forest residue was also fired. This fuel had higher ash content, considerably more sulphur and nitrogen compared to the wood pellets and wood briquettes.

Wood pellets (Table 8) are a refined and relatively homogenous biofuel with low moisture content. A wood pellet is a cylindrical fuel particle. The diameter and length of the pellets used in the Sandared boiler were 8 mm and 15-20 mm, respectively. For the tests in Sandared pellets of different densities were produced from the same raw material by Svensk Brikettenergi in Ulricehamn (Table 10). Wood briquettes of this study (Table 8) have a diameter and length of 75 mm and 50-100 mm, respectively. Briquettes of normal and low density (Table 10) were fired. Low-density briquettes disintegrate more easily and generate more fines, which affects the combustion conditions.

A combustion case using a mixture of 75 volume-% forest residue and 25 volume-% briquettes was also run. The forest residue was composed of branches and tops of trees, which were splintered and dried before combustion. In the domestic pellet burners, pellets of a diameter of 8 mm were used (Table 8). These were similar to the pellets fired in Sandared, both regarding size and chemical content. In the pellet stove, pellets of a diameter of 6 mm were fired. The chemical content was similar to the two preceding wood pellets, though the nitrogen content was slightly higher.

Table 8 Elemental analyses and heating values of the fuels. The numbers (except moisture) refer to dry fuel.

	Sandared, Reference Pellets	Pellet burner, Wood Pellets	Pellet stove, Wood Pellets	Fristad, Reference Briquettes	Fristad, Forest Residue
Moisture (wt-%)	7.7	7.6	7.1	8.8	15.8
Effective heating value (at constant pressure, MJ/kg)	19.12	18.98	19.01	18.80	19.20
Ashes (wt-%)	0.3	0.5	0.4	0.3	3.2
Sulphur (wt-%)	<0.01	<0.01	<0.01	<0.01	0.04
Carbon (wt-%)	50.6	50.2	50.4	49.9	50.8
Hydrogen (wt-%)	6.0	5.9	6.0	6.2	6.0
Nitrogen (wt-%)	0.08	0.08	0.11	0.09	0.66
Oxygen (wt-%)	43.1	43.2	43.0	43.4	39.4
Chlorine (wt-%)	<0.01	-	-	<0.01	0.02

Table 9 Chemical analyses of fuel ash. Fuel samples were ashed at 550 °C. The numbers refer to fuel ash.

	Sandared, Reference Pellets	Pellet burner, Wood Pellets	Fristad, Reference Briquettes	Fristad, Forest Residue
Main elements (weight-%)				
Aluminium, Al	0.64	2.23	0.41	2.73
Silicon, Si	2.98	11.4	2.32	15.9
Iron, Fe	0.60	1.24	0.38	1.16
Titanium, Ti	0.06	0.12	0.04	0.12
Manganese, Mn	3.71	2.13	3.54	2.18
Magnesium, Mg	3.59	2.71	3.38	2.36
Calcium, Ca	21.7	19.2	20.5	15.4
Barium, Ba	0.34	0.29	0.39	0.22
Sodium, Na	0.62	1.79	0.43	1.58
Potassium, K	9.50	7.74	12.8	6.28
Phosphorus, P	1.19	1.18	1.99	1.70
Trace elements (mg/kg)				
Copper, Cu	220	83	270	189
Vanadium, V	4	16	5	35
Chromium, Cr	150	72	43	73
Cobalt, Co	10	11	9	12
Nickel, Ni	25	27	24	50
Zinc, Zn	3200	2800	3500	2100
Lead, Pb	98	38	214	131
Cadmium, Cd	12	2.8	22	8
Arsenic, As	<20	<20	32	<20
Molybdenum, Mo	<10	<10	<10	<10

Table 10 The densities of the fuels.

Fuel	Bulk density (kg/m ³)	Piece density (kg/m ³)
Sandared, Low density pellets	530	1100
Sandared, Reference pellets	700	1200
Pellet burner, Wood Pellets	-	1200
Pellet stove, Wood Pellets	-	1200
Fristad, Low density briquettes	380	-
Fristad, Reference briquettes	480	930

Measurement Set-Up

The measurement set-up for a field measurement is presented in Figure 16. The principles for measurements in the laboratory were the same, but all runs did not contain analysis of all gases. The gas and particle sampling were performed in the flue-gas path downstream of the cyclone. As seen from the figure, there is three heated sampling lines: for gas, for dust, and for particle number and size analyses. The dashed lines indicate heating to a temperature of 180 °C (to avoid condensation). The concentrations of CO/CO₂, O₂, NO_x and THC (total hydrocarbons) were continuously measured with conventional instruments. Methane (CH₄), ethene (C₂H₄), ammonia (NH₃), water (H₂O), and in some cases benzene (C₆H₆) were analysed by FTIR (Fourier Transform Infrared Spectroscopy). The results of the FTIR-measurements are reported by Persson *et al.* (2001).

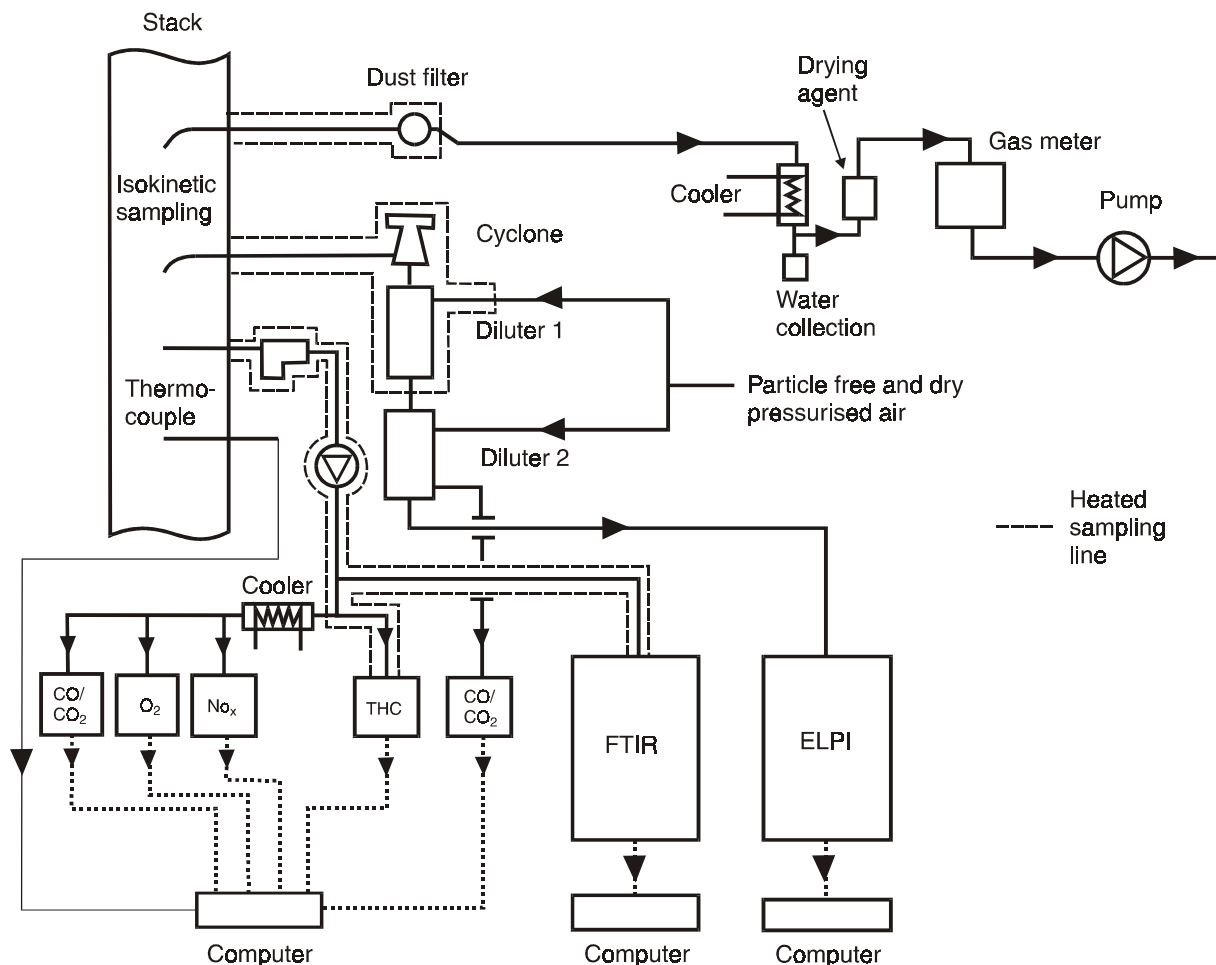


Figure 16 Measurement set-up during field measurements.

Particle Measurements

Particles were sampled in two separate lines, one for total mass concentration and another for number concentration and size distribution. The total mass of particulate emission, often called dust, was collected by isokinetic sampling and analysed gravimetrically. The method is simple; the dust is collected on a filter that is weighed before and after collecting dust. The moisture content of the flue gas was also determined gravimetrically by cooling and drying the gas. The precipitated water was then weighed and the mass increase of the drying agent was noted. As long as practically possible the dust measurements were performed according to Swedish standards (SS028426, 1991-12-04). According to standards, each measurement should be composed of dust sampling at four (the number is determined of the size of the flue gas channel and four is the lowest number) points in a cross section. The cross section where the measurement is carried out should have long enough section of straight channel upstream of and downstream of the cross section of the measurement. In the Sandared boiler one measurement point was used and there should have been somewhat longer sections of straight channel upstream of and downstream of the measurement point. In Fristad each measurement was represented of two measurements point in a cross section. The section of straight flue gas channel was sufficient in Fristad. The reason to deviations of standards was limitations in room and boiler design.

The fine particles originating from combustion were also sampled isokinetically through a separate sampling line. The flue gas was extracted and diluted in two steps. Two-stage dilution was applied to preserve the gas and particle conditions as much as possible, i.e. to avoid condensation and particle growth by coagulation. The flue gas was extracted through a heated probe and then passed a heated cyclone (180 °C). Thereafter it entered the first diluter, which was also heated to 180 °C. The second dilution was performed at room temperature. The dilution system is a two-step ejector diluter system, commercially available from Dekati Ltd. It gives a dilution ratio of about 1:50. As a control, the CO and/or CO₂ concentration was measured before and after dilution.

Number particle concentrations and size distributions were measured with an ELPI (Electrical Low-Pressure Impactor). Mass size distributions were obtained using a DLPI (Dekati Low-Pressure Impactor). Both instruments measured concentrations and size distributions in the size range 30 nm – 10 µm. The ELPI is more advanced and gives real-time data. Below, the basic mechanisms for inertial impaction, low-pressure impactors and the ELPI are given. For more detailed information the reader is referred to Hinds (1999) and/or Willeke and Baron (1993) and concerning earlier combustion measurements using an ELPI, see for example Moision (1999).

Inertial Impaction

An aerosol is passed through a nozzle and then the output stream is directed against a flat plate. This plate is called an impaction plate and forces the output stream to make a 90° bend. Particles with inertia higher than a certain value cannot follow the streamlines and collide, i.e. impact, on the plate. An ideal impactor operates so that all particles that are larger than a certain size are collected on the impactor plate. A few particles larger than the certain particle size corresponding to the particle stage, escape collection, and a few particles smaller than the given size are collected. The actual and ideal impactor curves are shown in Figure 17. The size in question is called the cutoff size, cutoff diameter, cut point, cutsize, cutoff, or d_{50} (Hinds, 1999). In practice most well designed impactors have a sharp cutoff size that is assumed to be ideal and their efficiency curves are characterised by a single number - Stk_{50} . This number is correlated to the Stokes number that gives 50 % collection efficiency.

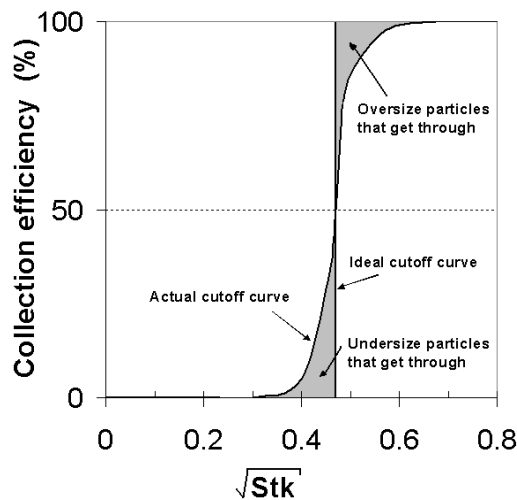


Figure 17 Actual and ideal cutoff curves of an impactor stage.

Low-Pressure Impactors

Low-pressure impactors measure mass size distributions of aerosols. They are designed according to the principle of inertial impaction and are advanced cascade-impactors (Hinds, 1999 and/or Willeke and Baron, 1993). The cascade-impactors are, which can be suspected by their name, composed of several single-stage impactors in series. The principle of a cascade impactor is shown in Figure 18. Each stage in the cascade-impactor represents a specific particle size range. This is accomplished by decreasing the nozzle diameter gradually in each step. An ideal cascade-impactor operates so that all particles smaller than the particles collected in the previous stage and larger than the cutoff size of the considered stage are collected. A cascade-impactor gives the mass size distribution during a certain time period of sampling. By preparing the impactor stages with weighed substrates (made of e.g. aluminium foil or polycarbonate foil) and weighing these substrates after collecting the particles, mass size distributions are obtained. The low-pressure impactor operates, as obvious from the name, at low pressures (and thus higher gas velocities) and can therefore capture smaller particle sizes than cascade-impactors. The low-pressure impactors are consequently a result of a development of the cascade-impactors. Particles are separated according to aerodynamic size in a low-pressure impactor. Particles may have different shape and density. A particle of an arbitrary shape and density has an aerodynamic diameter corresponding to a spherical particle with a density equal to water and the same sedimentation velocity as the arbitrary particle.

The stages of the low-pressure impactor (or the cascade-impactor) are described by their cutoff sizes. The cutoff size is related to the Stokes number that gives 50 % collection efficiency, also called $D_{50\%}$. The stages of the impactor are arranged in series, and consequently, several of the large particles that escape a certain stage impact on the next stage. That is why another way to characterise an impactor stage is also used, i.e. the geometric mean diameter. The geometric mean diameter of a certain impactor stage is equal to the square root of the product of the cutoff size of the stage and the stage above the stage concerned in the impactor. The difference between these diameters is illustrated in Table 11 where calibration data corresponding to the impactors used in this project are presented.

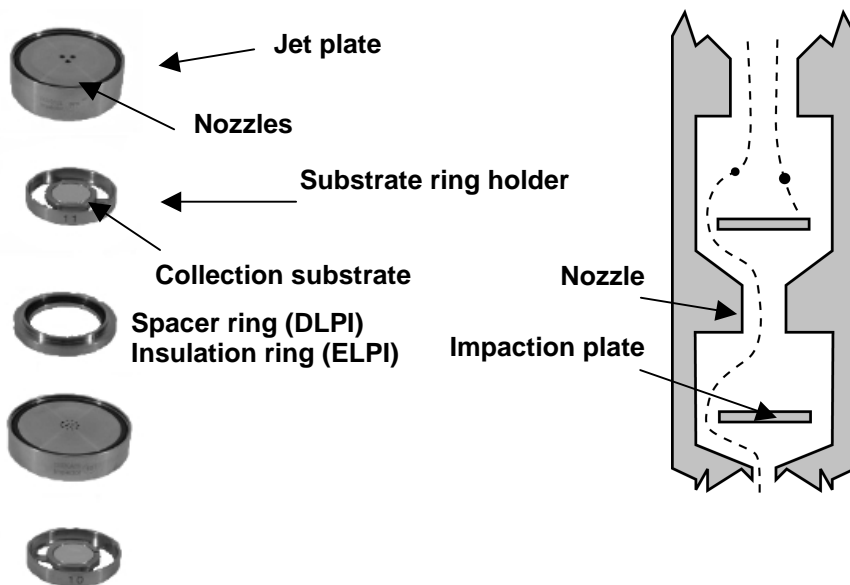


Figure 18 Photos of parts of the Dekati Low-Pressure Impactor to the left and a schematic picture of the principles of a cascade-impactor to the right.

Table 11 Calibration data of the impactors in the ELPI and DLPI used in this project.

Stage	1	2	3	4	5	6	7	8	9	10	11	12	13
ELPI													
D_{50%} (µm) ^a	0.029	0.059	0.103	0.166	0.255	0.393	0.637	0.99	1.61	2.46	3.97	6.69	10.15
D_i (µm) ^b	0.041	0.078	0.131	0.206	0.317	0.500	0.794	1.26	1.99	3.13	5.15	8.24	-
DLPI													
D_{50%} (µm) ^a	0.029	0.059	0.104	0.167	0.256	0.395	0.641	1.00	1.62	2.47	4.00	6.73	10.21
D_i (µm) ^b	0.041	0.078	0.132	0.207	0.318	0.503	0.801	1.27	2.00	3.14	5.19	8.29	-

^a D_{50%} means 50 % collection efficiency for particles of this aerodynamic diameter on this stage.

^b D_i means the geometric mean value, $D_{i, \text{stage } x} = (D_{50\%, \text{stage } x} * D_{50\%, \text{stage } x+1})^{0.5}$.

ELPI – Electrical Low-Pressure Impactor

Particle number concentrations and number size distributions, in the size range of 30 nm to 10 µm, were continuously measured using an ELPI (Electrical Low-Pressure Impactor). A schematic picture of the instrument is shown in Figure 19. The flue gas first reaches the ELPI's Corona charger, after which it enters a low-pressure impactor with several electrically isolated collection stages. The electrical current, carried by the charged particles into each impactor stage, is measured in real time by a sensitive multi-channel electrometer. The number concentration (number of particles at a normal cubic centimetre wet flue gas) measured with the ELPI is presented as PM₁₀. The particle sizes are measured as aerodynamic diameters.

Analyses of the composition of the particles

Qualitative analyses of the elemental composition of the submicron particles were performed by energy dispersive x-ray analysis (EDX). Investigations of the compounds were performed by time-of-flight secondary ion mass spectrometry (TOF-SIMS), described in Appendix 1. As alkali compounds are known to be important in the submicron particles, a new method

developed by Sjövall *et al.* (2001) was used to investigate the fractions of potassium sulphate, potassium chloride, potassium carbonate, and the corresponding compounds of sodium, that give rise to characteristic peaks in TOF-SIMS spectra. By TOF-SIMS spectra of reference samples with known quantities of these compounds, relative sensibility factors, corresponding to the characteristic peaks, were obtained. These sensibility factors were then used to determine the relative content of the different alkali compounds in the combustion samples.

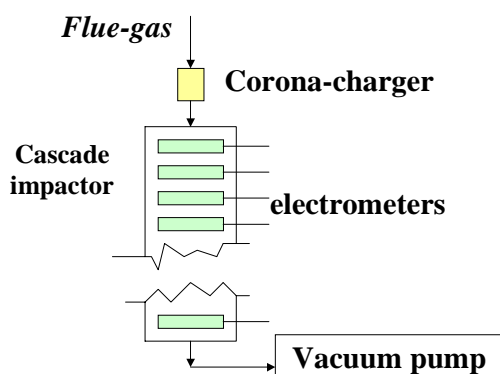


Figure 19 Schematic picture of the ELPI (Electrical Low-Pressure Impactor).

Gas measurements

Well-known gas analysis instruments were used as presented in Table 12. The instruments were calibrated before each measurement, and the measurement uncertainties were calculated according to the uncertainties of instruments and calibration gas and are shown in Table 13.

Table 12 Gas analysis instruments.

Gas analysis	Measurement principle	Trademark	Measurement range
CO/CO ₂	Non-dispersive infrared	Rosemount Binos 100	CO 0-500 ppm CO 0 – 3000 ppm CO 0 – 3 % CO ₂ 0 – 20 %
O ₂	Paramagnetic	M & C Instruments	0 – 100 % O ₂
NO _x	Chemiluminescence	ECO Physics	0 – 500 ppm
THC	Flame ionisation detector	J.U.M. Model 3-300A	1-1000 000 ppm

Table 13 Measurement uncertainties in gas measurements.

Measurement uncertainty	
O ₂	±0.2 %
CO ₂	±0.2 %
CO	±50 ppm
NO _x	±5 ppm
THC	±10 ppm
Dust	±10%

Measurement Plan

As a verification of the performance of the ELPI, a few simultaneous measurements with a SMPS (Scanning Mobility Particle Sizer) were performed. The SMPS measures number size distributions and concentrations according to the electrical mobility diameter. It is a well-established aerosol measurement instrument. For a description of the differential mobility analyser and the condensation particle counter, which are the main parts of the SMPS, the reader is referred to Hinds (1999) and/or Willeke and Baron (1993). In order to investigate the repeatability of measurements, the same particle measurements were repeated at a few occasions, uninstalling and installing the pellet stove in between.

The results from the parallel ELPI and SMPS (Scanning Mobility Particle Sizer) measurements are shown in Figure 20. SMPS measures electric mobility diameters whereas ELPI measures aerodynamic diameters. The electric mobility diameter is related to certain mobility in an electrical field. The aerodynamic diameter for a certain particle is defined as the diameter of a sphere of density 1 g/cm^3 with the same settling velocity as the particular particle. Particles of a certain Stokes diameter have the same settling velocity as spherical particles of the same density as the investigated particles. A density of two g/cm^3 of the emitted particles was assumed, and the results from the ELPI-measurement can be related to Stokes diameter, which is more similar to the electric mobility diameter. An assumed density of two g/cm^3 had a better agreement with the ELPI-measurements than using aerodynamic diameter. By knowing the true density of the particles, probably better agreement would have been obtained.

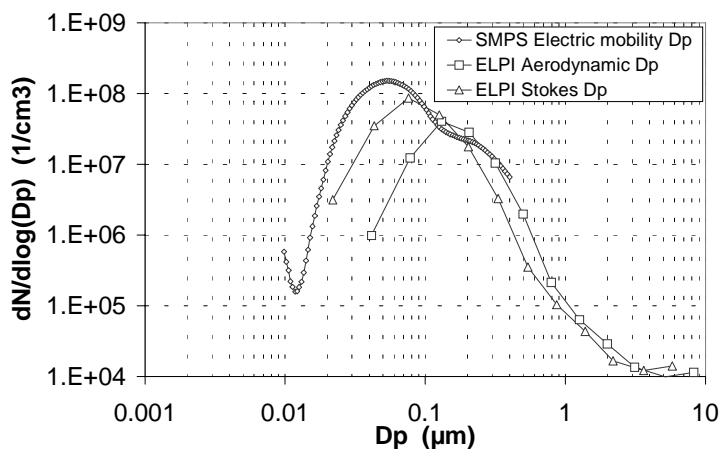


Figure 20 Particle number concentrations measured simultaneously with an ELPI and a SMPS (the curves are not normalised to any specific O_2 concentration).

Measurements of number concentrations and size distributions at a load of 100 % in the pellet stove were repeated at different occasions and shown in Figure 21 and Figure 22. The qualitative result, i.e. submicron particles dominate the emissions and the maximum around $0.13 \mu\text{m}$ is clearly repeated. The number concentration varied between $3.4 \cdot 10^7$ and $6.6 \cdot 10^7$ number of particles per cubic centimetre. The lowest value of total concentration deviates 30 % from the mean while the highest value deviates 40 % from the mean (as can be calculated from Table 18). However, the concentrations of the different sizes sometimes deviate more from mean (Table 14).

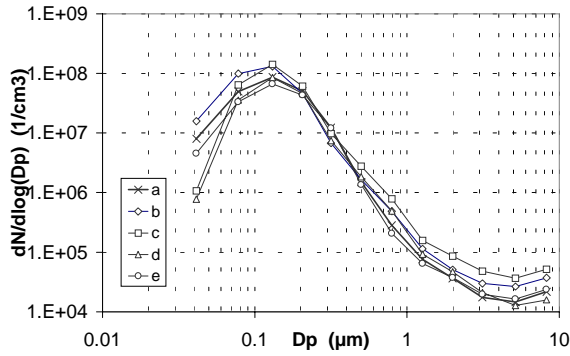


Figure 21 Number particle size distributions of particles emitted from the pellet stove at 100 % load at five occasions.

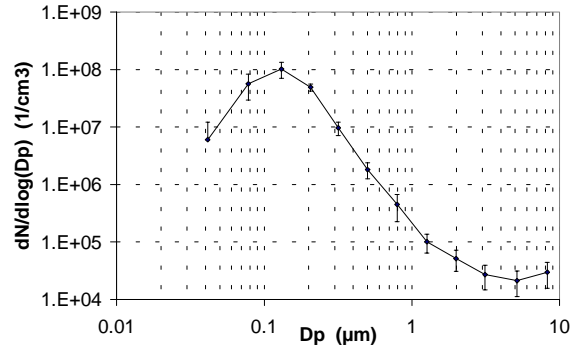


Figure 22 mean number particle size distributions of measurements a – e, standard deviations is marked as uncertainty columns.

Table 14 Mean values and lowest and highest values of number concentrations of different particle sizes are shown as well as the percentage deviation of the lowest/highest values from the mean value. Concentrations are divided by $dlog(Dp)$ as in Figure 21.

Dp (µm)	Mean (1/cm ³)	Case	Lowest (1/cm ³)	Lowest % deviation from mean	Case	Highest (1/cm ³)	Highest % deviation from mean	Range
0.041	6.04*10 ⁶	d	7.78*10 ⁵	87	b	1.58*10 ⁷	162	1.50*10 ⁷
0.078	5.64*10 ⁷	e	3.32*10 ⁷	41	b	9.93*10 ⁷	76	6.61*10 ⁷
0.131	1.02*10 ⁸	e	6.69*10 ⁷	35	c	1.41*10 ⁸	38	7.38*10 ⁷
0.206	4.93*10 ⁷	e	4.35*10 ⁷	12	c	6.08*10 ⁷	23	1.73*10 ⁷
0.317	9.68*10 ⁶	b	6.76*10 ⁶	30	a	1.23*10 ⁷	27	5.54*10 ⁶
0.500	1.82*10 ⁶	e	1.36*10 ⁶	25	c	2.77*10 ⁶	52	1.41*10 ⁶
0.794	4.50*10 ⁵	e	2.07*10 ⁵	54	c	7.84*10 ⁵	74	5.77*10 ⁵
1.26	1.01*10 ⁵	e	6.41*10 ⁴	36	c	1.57*10 ⁵	56	9.33*10 ⁴
1.99	5.15*10 ⁴	a	3.63*10 ⁴	29	c	8.62*10 ⁴	68	4.99*10 ⁴
3.13	2.71*10 ⁴	a	1.74*10 ⁴	36	c	4.78*10 ⁴	76	3.04*10 ⁴
5.15	2.13*10 ⁴	d	1.27*10 ⁴	40	c	3.66*10 ⁴	72	2.39*10 ⁴
8.24	2.99*10 ⁴	a	2.16*10 ⁴	28	c	2.97*10 ⁴	72	2.97*10 ⁴

District heating: The purpose of the present study was to characterise particle emissions, and therefore the measurements were performed downstream of the multicyclones in the district heating boilers. In the Sandared boiler, fired with wood pellets, the operation parameters varied were excess air, boiler load and density of pellets. In the Fristad boiler, the same operation parameters were varied, but also a case of firing a mixture of wood briquettes and dried forest residue was run. Mean values of emissions were measured during one hour or longer in each operation case. Before each run, the boilers were in stable operation.

Domestic heating: At the laboratory, emissions from domestic pellet burners were studied during continuous and intermittent operation. During continuous operation, measurements were performed at maximal thermal output and the burner was optimised for this case. Consequently, this case has the lowest possible emissions. In contrast, the normal operation of a domestic pellet burner is intermittent operation, adjusted to the heat demand of a house. The transients during intermittent operation were studied using the ELPI. During intermittent operation the boiler is set at thermostat operation. The pellet burner, then operating at

maximal thermal output, then heats the water in the boiler until the thermostat switches off. Then the burner fuel feed either turns off (burner with electric ignition) or switches to a very low level (burner with pilot flame) depending on the burner design. The water in the boiler is gradually cooled to a certain temperature. Then the thermostat switches on and the fuel feed starts again and also the combustion. This cyclic course is continuously repeated. A new cycle starts when the thermostat switches on and stops at next switch on. Intermittent measurements were performed at heat demands in a house corresponding to both 3 kW and 6 kW. The boiler was in thermal balance before each measurement case was initiated. The control devices of the test rig were adjusted according to a mean value of the water temperature of the boiler at $80\text{ °C} \pm 2\text{ °C}$. Each case was started and stopped at a switch of the boiler's thermostat, and each measurement case contained as a minimum two such cycles. For the case with the pellet stove, the influence on the emissions of the fuel feed rate and consequently of the thermal output was studied.

RESULTS

The particle emissions from a few small-scale wood pellet and briquettes combustor were characterised. The results from particle measurements and chemical analysis of particles are presented below.

Emission Characteristics of the Different Cases

Particle emissions were characterised by mass concentrations, and particle concentration as mean value during a time interval was obtained. In the district heating boilers the emissions fluctuated between 51 – 240 mg/m_n³ dry flue gas (Table 15, and Table 16). The pellet burners showed emission levels between 12 and 137 mg/m_n³. Maximal load in the pellet stove resulted in 65 mg/m_n³ (Table 17, and Table 18).

Table 15 Particle and gas emissions from the boiler in Sandared (normalised to 13 % CO₂). All numbers concern dry flue gas.

Load	O ₂ (%)	Fuel	Number conc., Dp=30 nm-10 µm (1/cm _n ³) ^b	Mass conc. (mg/m _n ³)	CO (mg/m _n ³)	THC ^c (mg/m _n ³)	NO _x ^d (mg/m _n ³)
20	7.5	reference	0.9*10 ⁷	96	14 853	268	35
20	8.5	reference	0.9*10 ⁷	115	11 618	298	48
20	9.9	reference	1.0*10 ⁷	78	96	3	52
50	4.7	reference	1.7*10 ⁷	69	9 197	144	53
50	7.4	reference	1.9*10 ⁷	74	1 053	13	69
50	9.2	reference	2.0*10 ⁷	97	3 414	25	58
100	3.7	reference	2.1*10 ⁷	71	299	1.0	78
100	6.8	reference	2.1*10 ⁷	51	55	<1	76
100^a	8.9	reference	3.4*10⁷	83	45	<1	83
100	9.3	low density pellets	3.5*10 ⁷	92	92	<1	83
100	9.5	ref. density 2 pellets	3.2*10 ⁷	83	44	<1	82

^a Extra bold type indicates the reference case, i.e. operation under normal conditions.

^b The emissions were calculated from wet to dry flue gas with the assistance of FTIR-data of water concentration in the flue gas.

^c THC (Total hydrocarbons) are represented as methane equivalents.

^d NO_x is presented as mg NO₂/ m_n³ dry flue gas.

In a few cases, the mass size distribution was determined. The mass maxima obtained were around 0.08 – 0.2 µm (Figure 23 - Figure 26). In the Fristad boiler a tendency of a second maximum around 2-3 µm was observed. There was also an indication of a supermicron mode during continuous operation of the pellet burner with pilot flame. In all combustion cases, the particle mass emissions were dominated by submicron particles.

Table 16 Particle and gas emissions from the boiler in Fristad (normalised to 13 % CO₂). All numbers concern dry flue gas.

Load (%)	O ₂ (%)	Fuel	Number conc., Dp=30 nm-10 µm (1/cm ³) ^b	Mass conc. (mg/m ³)	CO (mg/m ³)	THC ^d (mg/m ³)	NO _x ^e (mg/m ³)
20	9.6	reference	0.5*10 ⁷	83	1 569	1	70
50	7.4	reference	1.5*10 ⁷	58	609	2	63
100	3.9	reference	2.5*10 ⁷	68	552	4	82
100	4.8	reference	2.7*10 ⁷	72	224	<1	83
100^a	5.7	reference	3.6*10⁷	112	196	<1	85
100	7.5	reference	6.3*10 ⁷	240	265	4	93
100	6.3	low density	2.1*10 ⁷	87	560	2	72
100	6.5	forest residue and briquettes ^c	1.5*10 ⁷	117	93	<1	145

^a Extra bold type indicates the reference case, i.e. operation under normal conditions.

^b The emissions were calculated from wet to dry flue gas with the assistance of FTIR-data of water concentration in the flue gas.

^c 75 volume-% forest residue and 25 volume-% briquettes.

^d THC (Total hydrocarbons) are represented as methane equivalents.

^e NO_x is presented as mg NO₂/ m_n³ dry flue gas.

Table 17 Particle and gas emissions from the pellet burners, normalised to 10 % O₂. All results, except for number concentration that refer to wet gas, concern dry flue gas.

Burner	Case	O ₂ (%)	Number conc., Dp=30 nm-10 µm (1/cm ³)	Mass conc. (mg/m ³)	CO (mg/m ³)	THC ^c (mg/m ³)	NO _x ^d (mg/m ³)
Pilot flame ^a	100 % load	11.1	2.6*10 ⁷	47	76	6	156
Electric ignition ^a	100 % load	7.4	1.4*10 ⁷	34	245	5	148
Pilot flame	3 kW ^b	16.9	13.4*10 ⁷	132	1 951	370	136
Pilot flame	6 kW ^b	14.6	3.1*10 ⁷	12	733	115	146
Electric ignition	3 kW ^b	11.9	1.4*10 ⁷	33	244	14	140
Electric ignition	6 kW ^b	11.3	2.8*10 ⁷	137	2 053	87	132

^a The pellet burner with pilot flame was burning upwards, while the pellet burner with electric ignition was designed to burn forwards.

^b Intermittent operation.

^c THC (Total hydrocarbons) are represented as methane equivalents.

^d NO_x is presented as mg NO₂/ m_n³ dry flue gas.

Table 18 Particle and gas emissions from the pellet stove, normalised to 10 % O₂. All results, except for number concentration that refer to wet gas, concern dry flue gas.

Label	Load (%)	O ₂ (%)	Number conc., Dip=30 nm-10 µm (1/cm ³)	Mass conc. (Mg/m ³)	CO (Mg/m ³)	TIC ^a (Mg/m ³)	Kn _{ox} ^b (Mg/m ³)
a	100	13.8	4.5*10 ⁷	-	710	12	-
b	100	13.0	6.6*10 ⁷	-	540	12	-
c	100	12.9	5.9*10 ⁷	-	530	<1	-
d	100	13.5	3.7*10 ⁷	65	710	<1	-
e	100	12.8	3.4*10 ⁷	-	490	10	-
-	75	14.1	2.5*10 ⁷	-	500	9	140
-	50	15.5	3.7*10 ⁷	-	280	6	130
-	minimal	19.4	12.1*10 ⁷	-	4290	230	150

^a THC (Total hydrocarbons) are represented as methane equivalents.

^b NO_x is presented as mg NO₂/ m_n³ dry flue gas.

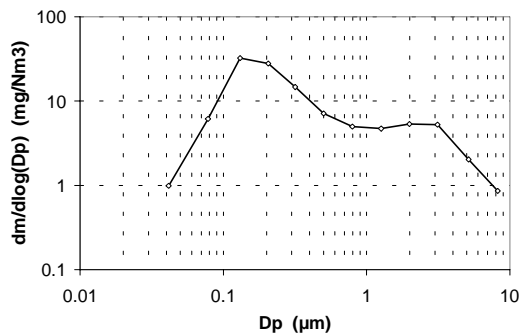


Figure 23 mass particle size distributions during combustion at the reference case at the boiler in Fristad (normalised to 13 % CO₂).

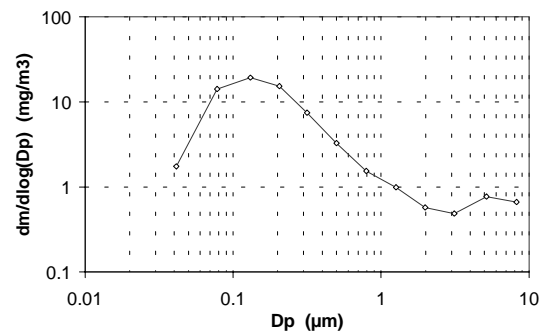


Figure 24 mass particle size distributions during continuously operation at the pellet burner with a pilot flame (normalised to 10 % O₂).

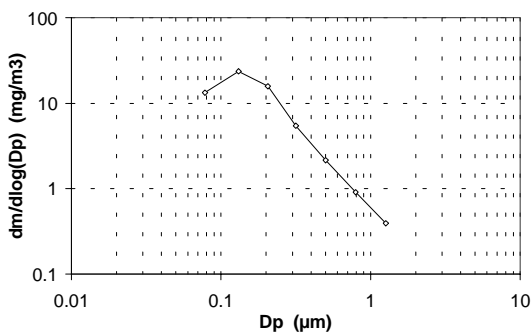


Figure 25 mass particle size distributions during continuous operation of the pellet burner with electric ignition (normalised to 10 % O₂). Please note that the weighing of particle fractions larger than 1 µm failed.

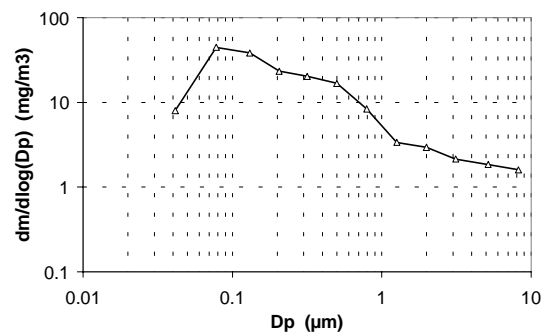


Figure 26 mass particle size distributions at 100 % load in the pellet stove (normalised to 10 % O₂).

Number concentrations in the emissions from the district heating boilers varied between $0.5 \cdot 10^7$ and $6.5 \cdot 10^7$ number of particles per normal cubic metre (Table 15, and Table 16), while those from the domestic heating devices varied between $1.4 \cdot 10^7$ and $13.4 \cdot 10^7$ number of particles per normal cubic metre (Table 17, and Table 18). The same instrument, ELPI, determined number size distribution simultaneously with number concentration. Maxima of number size distributions were between $0.080 \mu\text{m}$ and $0.3 \mu\text{m}$ (Figure 27 - Figure 35). From a number perspective the particle emissions were dominated by submicron particles.

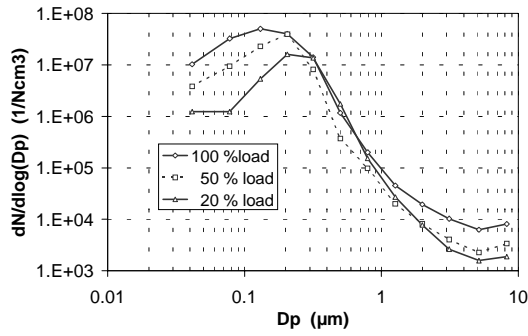


Figure 27 Number particle size distributions at different loads at the boiler in Sandared (the curves are normalised to 13 % CO₂).

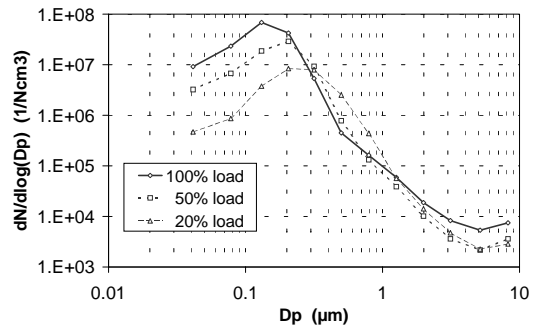


Figure 28 Number particle size-distributions at different loads at the boiler in Fristad (the particle concentrations are normalised to 13 % CO₂).

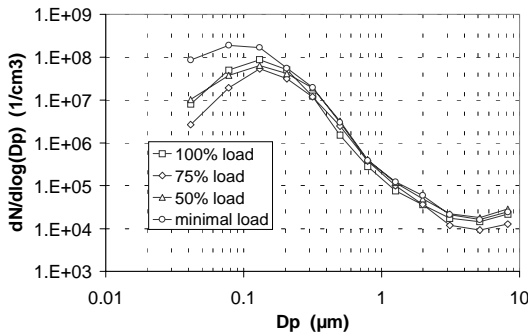


Figure 29 Number size distributions at 100 %, 75 %, 50 % and minimal load at the pellet stove. The curves are normalized to 10 % O₂.

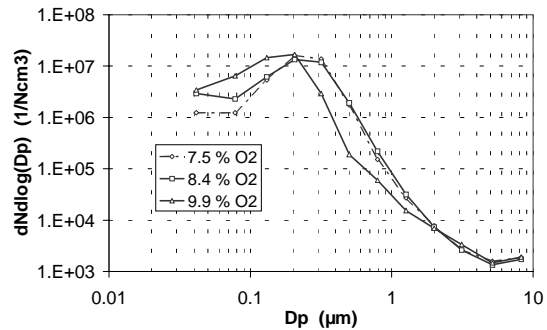


Figure 30 Number particle size distributions at 20% load for different excess air ratios at the boiler in Sandared (the curves are normalised to 13 % CO₂).

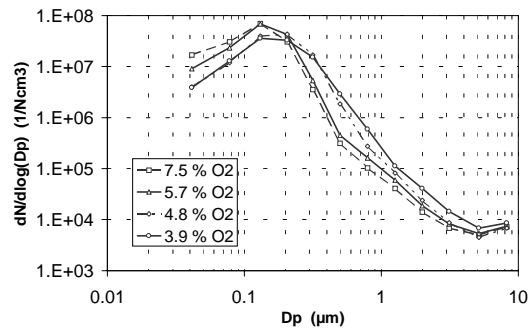


Figure 31 Number particle size distributions at 100% load and different excess airs at the boiler in Fristad. The distribution curves are normalised to 13 %C O₂.

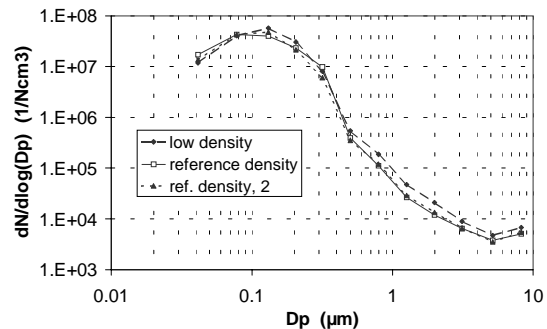


Figure 32 Number particle size distributions at different pellet densities at the boiler in Sandared. The distribution curves are normalised to 13 %C O₂.

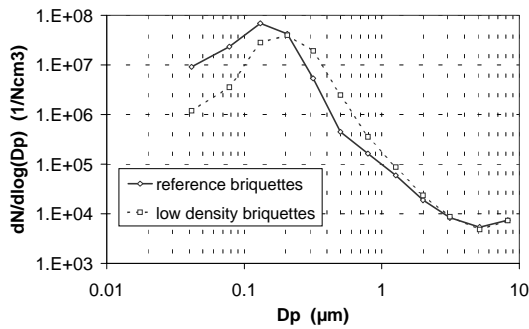


Figure 33 Number particle size distributions at different briquette densities at the boiler in Fristad (the curves are normalised to 13 % CO₂).

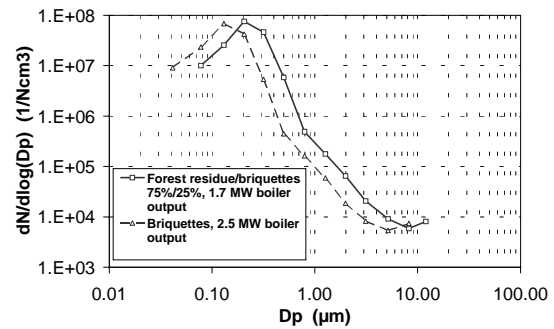


Figure 34 Number particle size distributions during combustion at the reference case at the boiler in Fristad compared to combustion of a mixture of 75 volume-% forest residue and 25 volume-% briquettes.

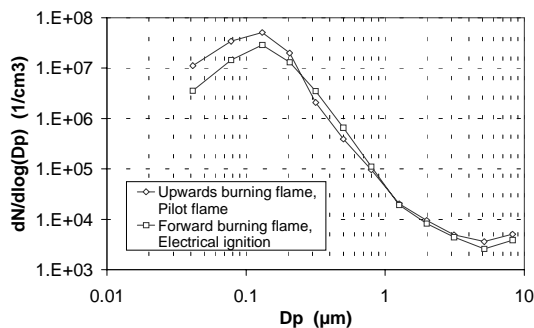


Figure 35 Number particle size distributions during continuous combustion at pellet burner with pilot flame and electric ignition respectively.

The Influence of Operating conditions and Fuel Quality

The combustion conditions vary as a domestic burner turns on and off, and there are also differences between burner designs. Under favourable combustion conditions, the particles consist mainly of ash. Favourable combustion leads to small emissions of hydrocarbons, because the organic material burns out. Transient courses in the pellet burner with pilot flame were studied by measurements of number concentration of particles and concentration of total hydrocarbons. Total hydrocarbons were measured as an indicator of combustion conditions. Large emissions of hydrocarbons indicate unsatisfactory combustion conditions and probably soot emissions. In Figure 36 emitted particles and total hydrocarbons from the burner seem to produce frequent peaks. The peaks correspond to the period when the pellet burner is turned off, but the glowing bed of fuel in the burner cup continues to burn. When the burner was in steady operation (i.e. turned on), both particle and hydrocarbon emissions were low.

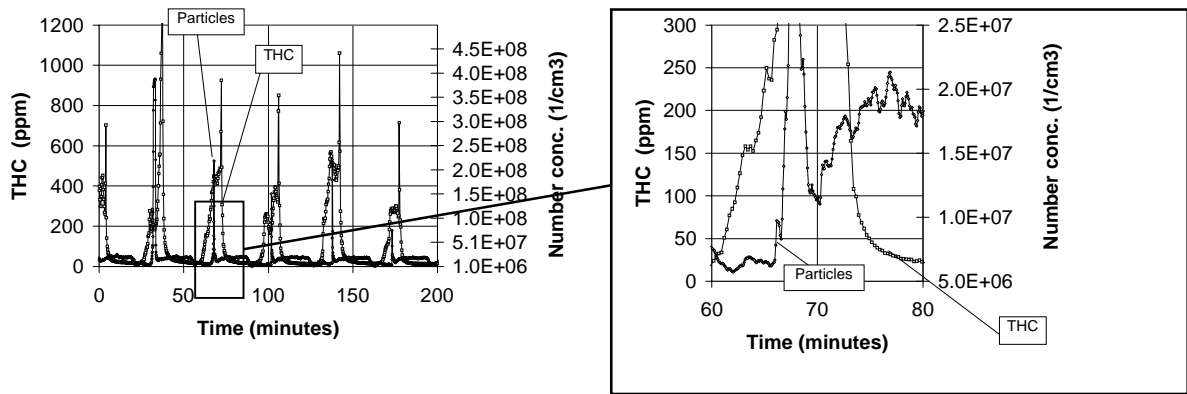


Figure 36 Particle and total hydrocarbon transient during combustion in the pellet burner with pilot flame and at a heat demand of 6 kW.

The influence of operation parameters on the particle emission was investigated. In the district heating boilers, the parameters were boiler load, excess air and fuel density. In the Fristad boiler, a case of co-combustion with briquettes and dried forest residue was also included. Forest residue was chosen because of high ash content. In the laboratory, the influence of load was investigated in the pellet stove. In the Sandared boiler, there was a slight tendency to an increase in particle emissions at decreasing load (Figure 37). No clear relation between particle emissions and level of excess air was seen (Figure 37). In the Fristad boiler, the four measurements at 100 % load indicate that particle emissions increased somewhat with excess air (Figure 38). There was just one measurement at 20 % and 50 % load and no relation between particle emissions and load was established. Minimum load in the pellet stove led to emissions of considerably larger number of particles than at remaining loads (Figure 39). This may be a result of a considerably higher level of excess air (and thus higher gas flows) than at the higher loads.

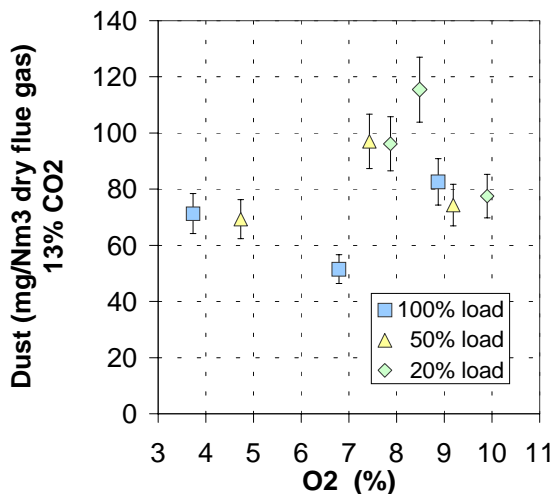


Figure 37 Dust emissions at different boiler loads and excess air ratios during pellet combustion in the Sandared boiler. Measurement uncertainties are illustrated by bars.

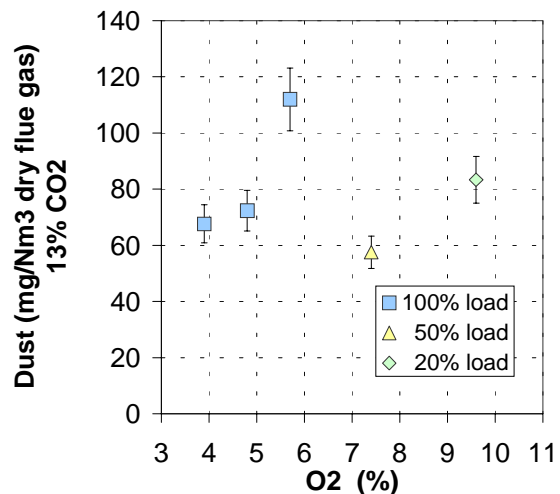


Figure 38 Dust emissions at different boiler loads and excess air ratios during briquette combustion in the Fristad boiler. Measurement uncertainties are illustrated by bars.

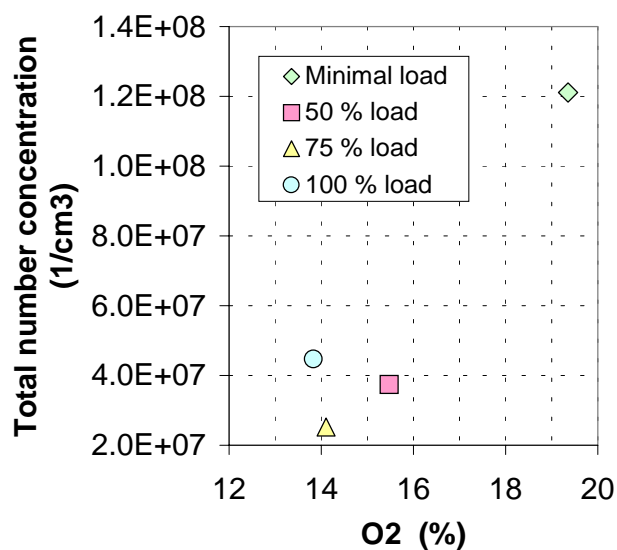


Figure 39 Particle number concentrations at different loads in the pellet stove. The emissions are normalised to 10 % O₂ and corresponding O₂-concentrations are indicated on the horizontal axis.

The maximum of the number size distribution tended to shift to larger particle sizes at low excess air and low load (Figure 27 - Figure 31). Combustion of pellets of different densities caused no significant differences in number size distributions (Figure 32 - Figure 33). However, combustion of low-density briquettes caused a maximum at higher particle size than the reference briquettes. This was also the case during combustion of a mixture of forest residue and briquettes (Figure 34). Combustion of low-density briquettes led to lower particle emissions than combustion of reference briquettes and combustion of the mixture of forest residue and briquettes caused emissions of the same concentration level as the reference fuel.

Analyses of the Particle Composition

Finally, the chemical content of the particle emissions was determined. Number and mass size distributions showed that particle emissions were dominated by submicron particles. Consequently, chemical analyses were performed on some fractions of the submicron particles. The main components of the particles were determined by EDX-analysis, and were potassium, sulphur, chlorine and oxygen. The samples also contained small amounts of sodium, magnesium and zinc. The distribution between potassium sulphate, potassium chloride, potassium carbonate, and the corresponding sodium compounds was determined by using a TOF-SIMS instrument. The dominant alkali compound was potassium sulphate, and potassium chloride was the second most important compound (Figure 40-Figure 49). Note that Figure 40-Figure 49 consider mass fractions.

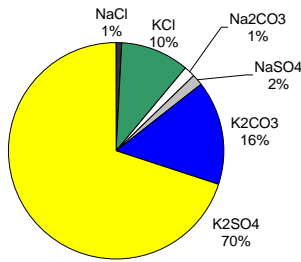


Figure 40 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.13 μm . The sample was collected from combustion of pellets in Sandared (reference case).

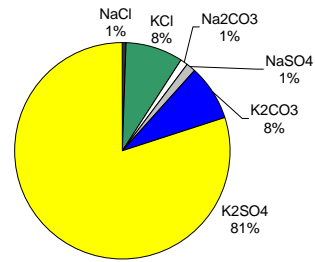


Figure 41 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.32 μm . The sample was collected from combustion of pellets in Sandared (reference case).

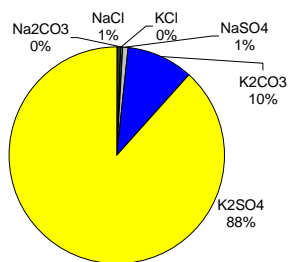


Figure 42 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.13 μm . The sample was collected from combustion of briquettes in Fristad (reference case).

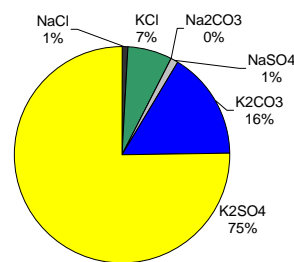


Figure 43 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.32 μm . The sample was collected from combustion of briquettes in Fristad (reference case).

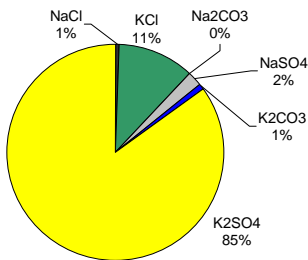


Figure 44 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.13 μm . The sample was collected from combustion of forest residue in Fristad.

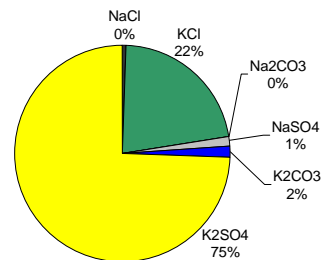


Figure 45 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.32 μm . The sample was collected from combustion of forest residue in Fristad.

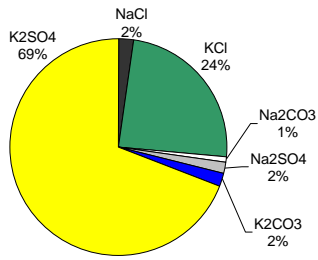


Figure 46 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.13 μm . The sample was collected from combustion of wood pellets in the pellet burner with electric ignition.

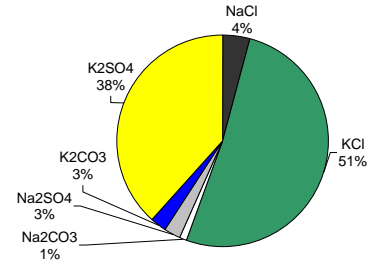


Figure 47 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.32 μm . The sample was collected from combustion of wood pellets in the pellet burner with electric ignition.

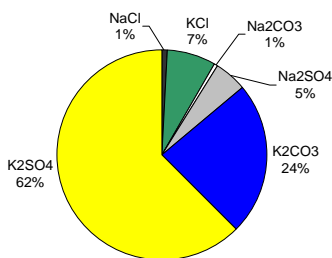


Figure 48 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.13 μm . The sample was collected from combustion of wood pellets in the pellet stove.

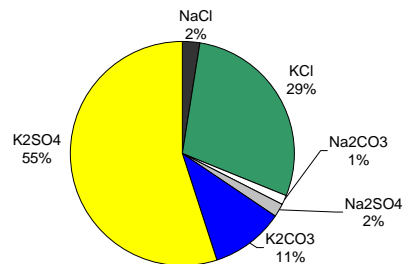


Figure 49 Alkali speciation for a particle fraction collected on an impactor plate with a geometric mean diameter of 0.32 μm . The sample was collected from combustion of wood pellets in the pellet stove.

DISCUSSION

This study presents emission data from combustion of wood pellets and wood briquettes in commercial domestic combustion devices and smaller district heating plants. The particle emissions are characterised by mass and number concentrations, size distributions and the inorganic composition of the particles. Transients, measured by the ELPI, and the influence of operation parameters on the particle emissions were also studied.

Mass, Number Concentrations, and Size Distributions

The mass concentrations of particles from combustion of wood pellets and wood briquettes were between 12 and 240 mg/m^3 . The lowest mass concentration was obtained during intermittent operation, at 6 kW boiler output, of the pellet burner with pilot flame. The highest mass concentration occurred during combustion of wood briquettes at high excess air ratio in the Fristad district heating boiler. In domestic devices, the combustion condition significantly affects the mass concentration of particles emitted. Unsatisfactory combustion conditions can lead to large emissions, see for example Muhlbaler Dasch, (1982), and McDonald *et al.*, (2000). The concentrations of carbon monoxide and total hydrocarbons can be regarded as indicators of the completeness of combustion. In Figure 50 and Figure 51 the flue gas concentrations of carbon monoxide and total hydrocarbons are plotted against the mass concentration of emitted particles. The data are taken from the pellet burner cases, and the mass concentration of particles tends to increase with CO and THC concentrations, as expected.

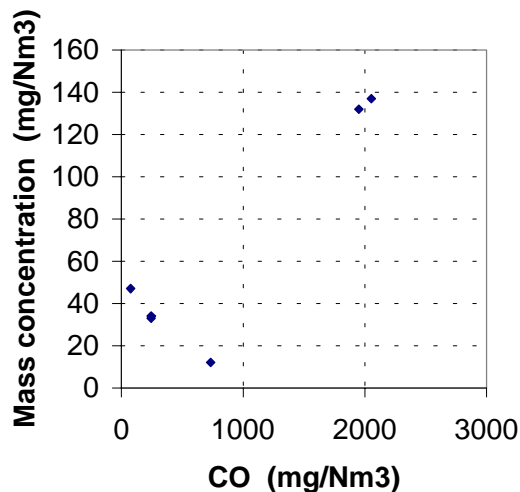


Figure 50 The CO concentration in the flue gas versus the mass concentration of particles for combustion in a pellet burner.

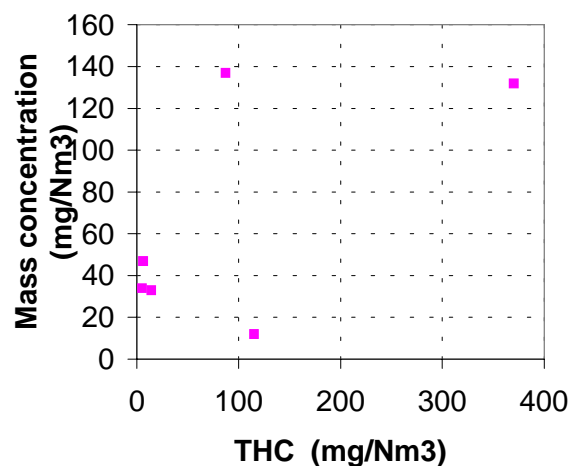


Figure 51 The concentration of total hydrocarbon in the flue gas versus the mass concentration of particles for combustion in a pellet burner.

The highest mass concentration of particles measured in this study was at 100 % load and highest excess air ratio in a grate-fired boiler (2.5 MW thermal output). This may indicate that a large fraction of the bottom ash particles follows the gas flow because of the high gas velocity in this case. Another possible explanation is unsatisfactory function of the multicyclone for this case.

The submicron mode is always present in mass and number size distributions during combustion of biomass. In certain cases, there is also a supermicron particle mode present. Nussbaumer and Hasler (1999) stated that particularly combustion of bark caused a bi-modal size distribution in particle emissions from domestic combustion with a particle size around 5 μm . Studies of the particle emissions upstream of the dust-separation device in biomass-fired district heating boilers (Lind *et al.*, 1997, Valmari *et al.*, 1998, and Lind *et al.*, 1999) show bi-modal mass size distributions. The previous measurements showed one submicron and one supermicron maximum in the mass size distribution. However, there are studies of smaller grate-fired district heating boilers only indicating the submicron mass mode (Kaufmann and Nussbaumer, 1998, Machan, 1998, Obenberger *et al.*, 2001). The supermicron maxima reported are in the range 1 – 100 μm . Many studies of particle mass size distributions are carried through with instruments with the higher detection limit around 10 μm , as a consequence information about particles larger than 10 μm are missing (and sometimes supermicron maxima). In boilers that are larger than those for domestic heating, the flue gas flows velocities are higher, and consequently more of the supermicron particles follow the flue gas stream out from the furnace. Depending on the efficiency of the dust separation device, particles are removed in a higher or lower degree. The present study of particle emissions from district heating boilers was carried out downstream of the boilers' multi-cyclones. Most of the supermicron particles should be removed from the gas (Appendix 2), whereas the submicron particles pass through the cyclone. A sufficient number of particles escape the cyclone to give a slight supermicron mass mode, and thus a mode of large particles must be present upstream of the cyclones. The number size distributions in the flue gas from the district heating plants, pellet burners and the pellets stove were uni-modal with maxima in the submicron range. The mass size distribution of the particle emissions from the pellet burner with pilot flame also indicates a supermicron mode, but no supermicron mode was observed in the particle emissions from the pellet stove. There are a few reports about the mass size distribution in relation to residential heating. Muhlbaler Dasch (1982) has reported that wood combustion leads to particle emissions where most of the mass is of submicron size. This agrees with the experiments of Rau (1989) for high excess air ratios, but he also reported bi-modal mass size distributions at low excess air ratios. Oser *et al.* (2001) reported bi-modal mass size distribution in the particle emissions from a large domestic combustor device, a 120 kW wood chips burner, during combustion of wet wood chips. The present residential measurements consider combustion of dry bio-fuels (less than 10 % moisture) at sufficient supply of air. Consequently, the mass size distributions in this work agree with previous results obtained domestic combustion under favourable combustion conditions, i.e. they show unimodal size distributions.

The Influence of Operating Conditions and Fuel Quality

The results show displacements in the number size distribution towards larger particle sizes at lower boiler load and lower excess air ratio. Lower load or lower excess air ratio often mean a larger degree of unsatisfactory combustion conditions, which means that both more organic condensation nuclei, as well as condensable gas might be present. These conditions favour nucleation and surface growth by condensation. In addition, low load and low excess air ratio are typically related to lower gas flow and thus higher residence time. High residence time favours particle growth by coagulation. The processes behind the emitted particle size distributions are thus a combination of mechanisms.

Combustion of wood briquettes of low density or a mixture of 75 volume-% dried forest residue and 25 volume-% wood briquettes also leads to a displacement of the number mode

towards larger particle sizes compared to normal-density briquettes. There was also a tendency of lower number concentration as the maximum in the size distribution was displaced towards larger particle sizes. These results are in agreement with Wieser and Gaegauf (2000), and Gaegauf *et al.* (2001), who showed that fewer particles are emitted at lower excess air and also the particle mode was displaced towards larger sizes. Kaufmann (1997) also reported the typical particle size to increase at lower excess air ratios. However, others have received contrary results. Oser *et al.* (2001), showed that increased air supply leads to size distributions displaced towards smaller sizes, and Hueglin *et al.* (1997) stated that a reduction in the air supply, as well as too high an excess air ratio, leads to an increase in the particle size and in the total number concentration. One explanation is that burner /boiler design affects such relations, and also the interval of excess air levels is important. In Figure 52 and Figure 53 the present data work are compared with literature values, the same literature data as in the introduction section is used. No clear trends are observed, of the influence of oxygen on mass or on number concentrations. For combustion of wood logs in a stove, Hueglin *et al.* (1997) and Gaegauf *et al.* (2001) reported that for the emissions of particles, the number concentration and size distribution mode strongly depend on the phase of combustion. The number maximum attained its largest particle size during start-up phase and decreased to a smaller size during the intermediate phase and to the smallest size during the burn-out phase. In the present results the influence of fuel density on the particle emissions is different in the cases of pellets and briquettes. Combustion of low-density briquettes displaced the number distribution towards larger particle size compared with the combustion case with reference briquettes. Low-density fuel disintegrates easier and may be compared with a fuel containing larger amounts of fines. With a larger number of fines, part the combustion is displaced downstream of the fuel bed. Thus, with a larger quantity of fines the total dust emissions may increase as well as the emissions of unburnt. On the other hand, combustion of wood pellets of different densities gave no significant displacement of the number size distribution. Oser *et al.* (2001) have reported that an increased amount of fines in the fuel leads to an insignificant increase of the particle emissions, which is in agreement with the present results on wood pellets. The difference in results may be caused by the difference between the two briquette qualities, which was considerably larger than between the different wood pellets and the different fuels fired by Oser *et al.* (2001).

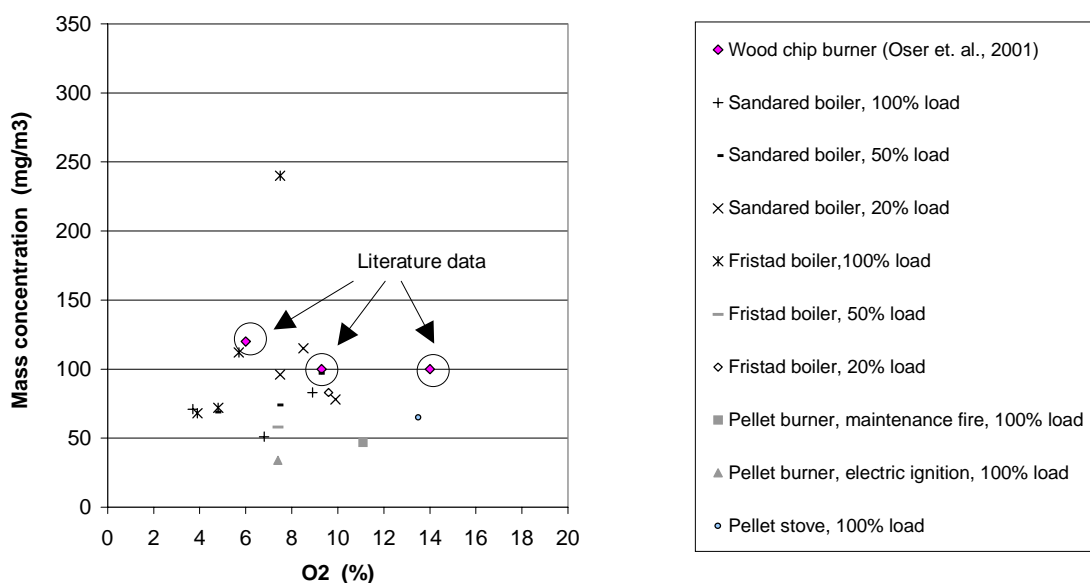


Figure 52 Oxygen concentrations in the flue versus the mass concentration of particles emitted. The present results are shown together with literature data.

The mass concentrations of the particles emitted from pellet burner combustion were about in the same range as earlier investigations (Wieser and Gaegauf, 2000, Gaegauf *et al.*, 2001). No data on mass concentrations of emitted particles from grate combustion of wood pellets and wood briquettes were found in the literature. However, results on combustion of wood-chips in a smaller grate-fired boiler have been reported to give particle emissions of about the same level as in the present study (Obenberger *et al.*, 2001). No obvious trend was found regarding the influence of moisture content on mass and number concentration, as shown in Figure 54 where results from the present investigation are compared with literature data. Also, there was no trend found for moisture content and number concentration, Figure 55.

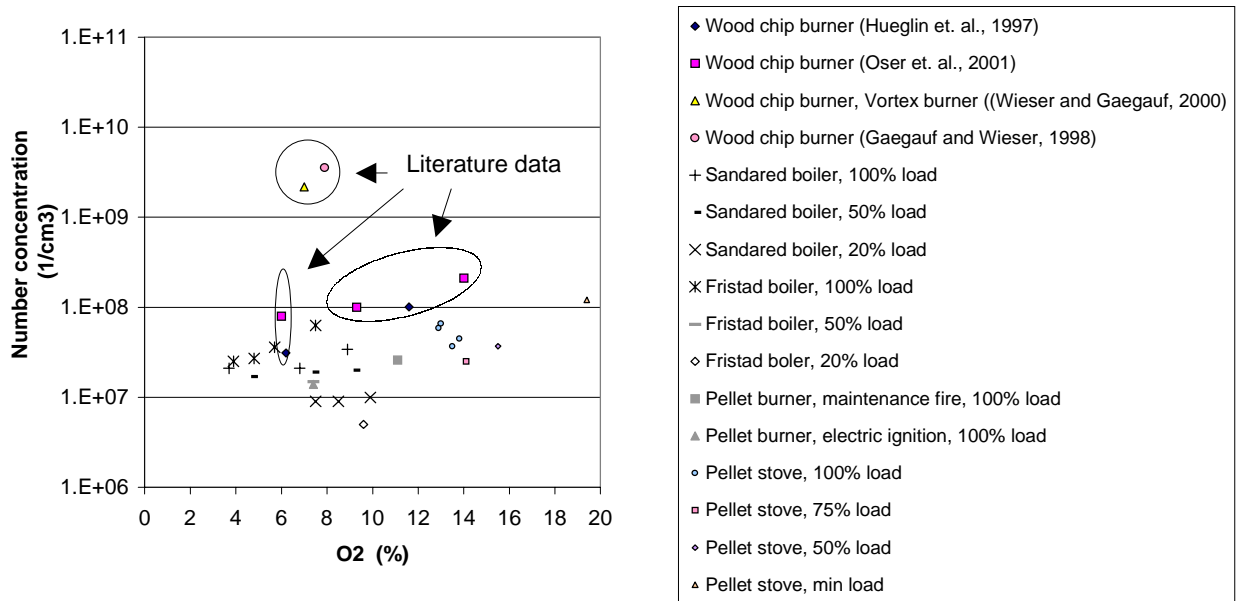


Figure 53 Oxygen concentrations in the flue versus the number concentration of particles emitted. The present results are shown together with literature data.

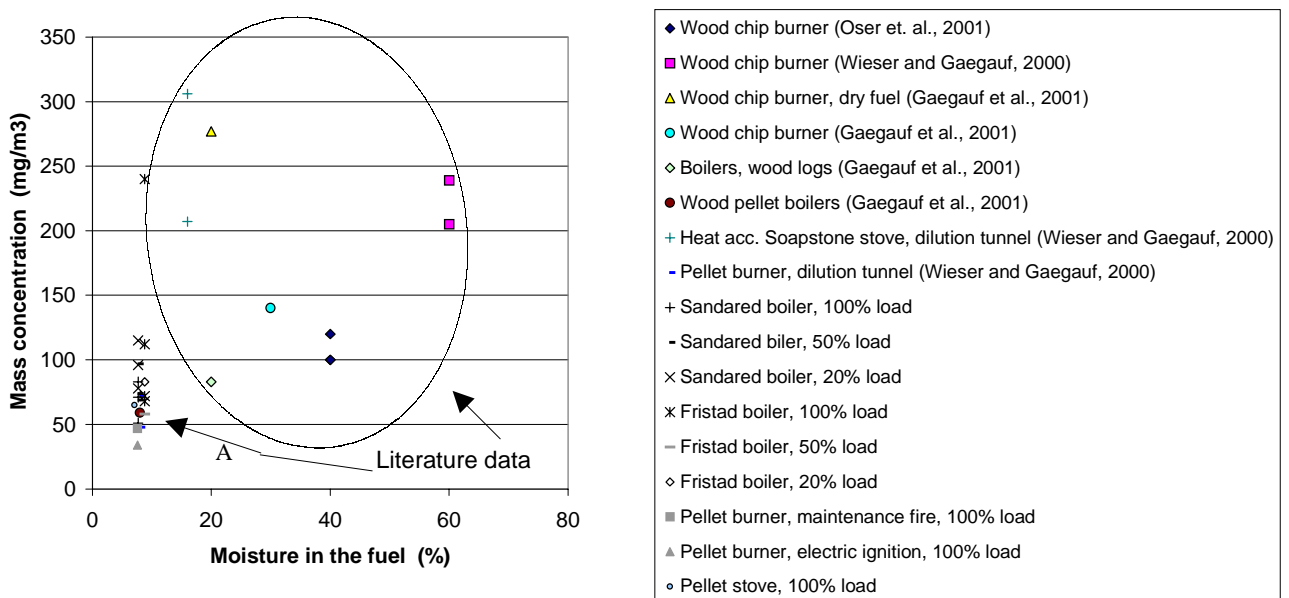


Figure 54 Moisture content in the fuel versus the mass concentration of particles emitted. The present results are shown together with literature data. The arrow labelled A indicates a single literature value behind the present data, wood pellet boilers (Gaegauf *et al.*, 2001).

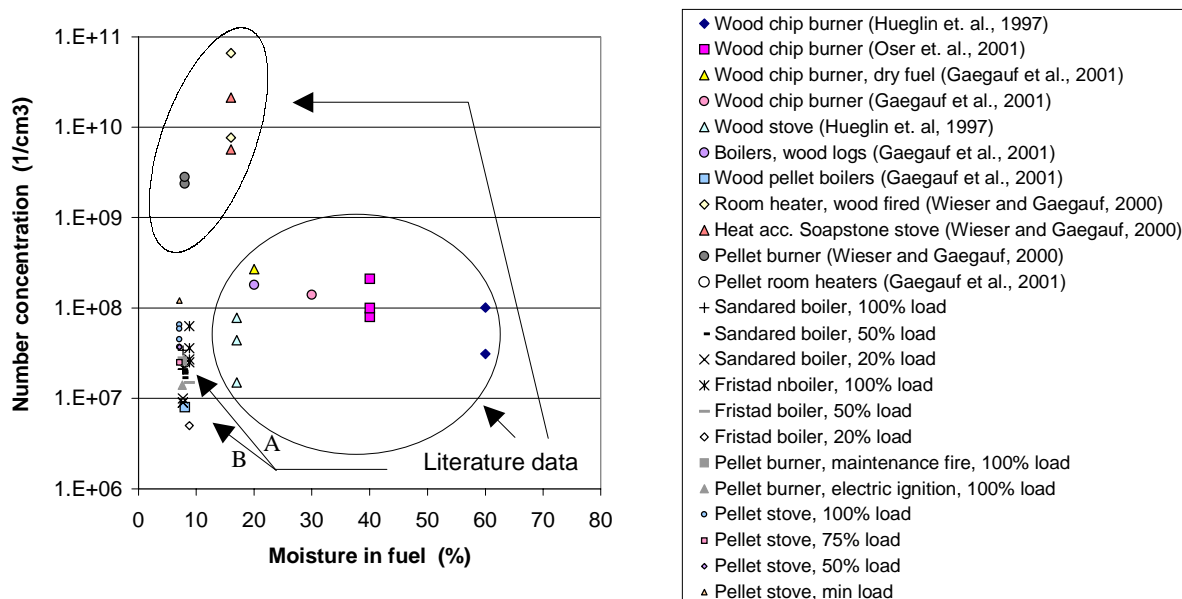


Figure 55 Moisture content in the fuel versus the number concentration of particles emitted. The present results are shown together with literature data. The arrow labelled A indicates a single literature value behind the present data, pellet room heaters (Gaegauf et al., 2001). The arrow labelled B is pointing at a literature data near the present data, wood pellet boilers (Gaegauf et al., 2001).

In the study of the transient behaviour of a pellet burner it was observed that the peaks in particle emissions follow the peaks in total hydrocarbons (Figure 36). This indicates the presence of particles from incomplete combustion, probably soot, which supports the assumption that unsatisfactory combustion conditions lead to increased particle emissions.

Analyses of The Particle Composition

The present observation that potassium sulphate is the main compound in the emitted submicron particles deviates somewhat from earlier results, which have reported potassium chloride as the main compound, and with potassium sulphate only second in importance (Lind *et al.*, 1999, Kaufmann and Nussbaumer, 1998, Lind *et al.*, 1999, Christensen, 1995, Kaufmann and Nussbaumer, 1998, Machan, 1998, Brunner *et al.*, 2000). The fuel composition and content of inorganic ash-forming matter varies. Previous studies of chemical composition of particles from biomass combustion focus on forest residue, hay and straw. Forest residue, hay and straw normally contain larger amounts of ash, and larger concentration of chlorine and sulphur, than wood pellets and wood briquettes. These fuels also contain larger amounts of water than the refined fuels, pellets and briquettes. For woody fuel, which has a relatively low alkali content as well as a low chlorine concentration, Dayton and Milne (1996) proposed that alkali compounds primary are released through vaporisation or decomposition of alkali sulphate. During combustion of fuel with a high concentration of alkali as well as chlorine, for example herbaceous fuel, grass, and straw, Dayton and Milne (1996) on the other hand suggested that alkali metal chlorides are the primary species released. The current investigated fuels have low concentrations of alkali, chlorine and sulphur, and the main alkali compound found in the current fly-ash was potassium sulphate. Thus literature data and the present study imply that chlorine to sulphur ratio is very important for the composition of the emitted submicron particles. However, it is clear that more knowledge about the mechanisms of alkali release and the subsequent formation of particles is needed.

CONCLUSIONS

Particle emissions from small-scale combustion of wood pellets and wood briquettes have been characterised and the following conclusions can be made:

- Domestic combustion of biomass typically leads to particle emissions dominated by submicron particles, and with uni-modal size distributions, both for mass and number concentrations. This is shown for combustion of wood pellets in this study and it confirms earlier work. Particle emissions downstream multi-cyclones are dominated by submicron particles, both for mass and number concentrations. There is a potential to reduce the particle emissions from small-scale district heating by installing additional dust separation devices.
- The mass concentration of particles increases during unsatisfactory combustion conditions due to particles originating from incomplete combustion. These include both soot and condensable organic particles. Unsatisfactory combustion conditions can result from low combustion rate due to high moisture content in the fuel or low excess air ratio.
- The excess air affects the number size distribution. The number maximum is at the largest particle size during lowest excess air ratio, and at the smallest size during highest excess air ratio, for the combustion cases investigated. Combustion of wood briquettes of low density or a mixture of 75 volume-% dried forest residue and 25 volume-% wood briquettes also leads to a displacement of the number mode towards larger particle sizes compared to normal-density briquettes.
- In this investigation potassium sulphate was found to be the main compound in the emitted submicron particles, and potassium chloride to be the second most important, but in earlier field studies potassium chloride has been observed as the main component. A possible explanation can be found in the difference in chlorine to sulphur ratio. In the previous studies, the chlorine content was higher in the fuels investigated (forest residue, hay, straw) than in the present investigation of pellets and briquettes.
- The study provides a starting point for investigations of the ash-forming mechanisms related to combustion of wood pellets and wood briquettes. Further studies should give a more complete picture of the ash emissions and ash forming steps during combustion of wood pellets and wood briquettes.

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APPENDIX 1

DESCRIPTION OF THE TOF-SIMS ANALYSIS

A brief description of the time-of-flight secondary ion mass spectrometer (TOF-SIMS) technique is given.

The Analysis Technique

SIMS

SIMS (secondary ion mass spectrometry) is a method for analysing the elemental and chemical composition of materials, in particular their surfaces. The general principle of SIMS analysis is briefly as follows. The material is irradiated with a focused beam of almost monoenergetic ions (primary ions). The primary ions penetrate the material and transfer their kinetic energy to the target atoms via a collision cascade. As a result of the collision cascade, atoms, clusters and molecules are emitted from the material (so called sputtering). A fraction of the sputtered particles are ionized (secondary ions) and therefore possible to analyse with respect to mass to charge ratio. The secondary ion mass spectrum gives information about the elemental and chemical composition of the material. Since the secondary ions originate only from the 1-5 outermost atomic/molecular layers of the material, the technique is highly surface sensitive.

TOF Mass Spectrometry

In a TOF-SIMS instrument, the secondary ion masses are measured by a time-of-flight (TOF) measurement. The technique is based on pulsed primary ion beams. The secondary ions generated by each primary ion pulse are accelerated to a constant kinetic energy, and then fly through a field free space before they hit a detector, where their intensity is measured as a function of flight time. Since ions with different masses have different velocities at a given kinetic energy, the measured flight times of the ions can easily be converted to their masses. The advantage of TOF mass spectrometry, compared to other mass spectrometry techniques (quadrupoles and magnetic sectors) is that it offers a unique combination of wide mass range, high mass resolution and high transmission.

Analysis Modes

TOF-SIMS allows several different analysis modes. Spectrometry can be done either in the static mode or the dynamic mode. In *static SIMS*, low primary ion beam doses are used and the mass spectrum is measured before a significant part of the surface layer has been chemically modified or sputtered away. In *dynamic SIMS*, higher primary ion intensities are used, which means that the surface is etched away layer by layer and that the analysis is done at a continuously increasing depth from the original surface (*depth profiling*). Dynamic SIMS therefore gives information about the depth distribution of elements. The use of focused beams also allows *analysis of small areas* (e.g. particles or fibres), and by rastering the primary ion beam and recording spectra from each analysis point, the lateral distribution of different elements and compounds at the surface can be visualized (*ion imaging*). Ion imaging can also be combined with depth profiling, which means that the three-dimensional distribution of elements and compounds can be analysed.

APPENDIX 2

SEPARATION CHARACTERISTICS OF MULTICYCLONES

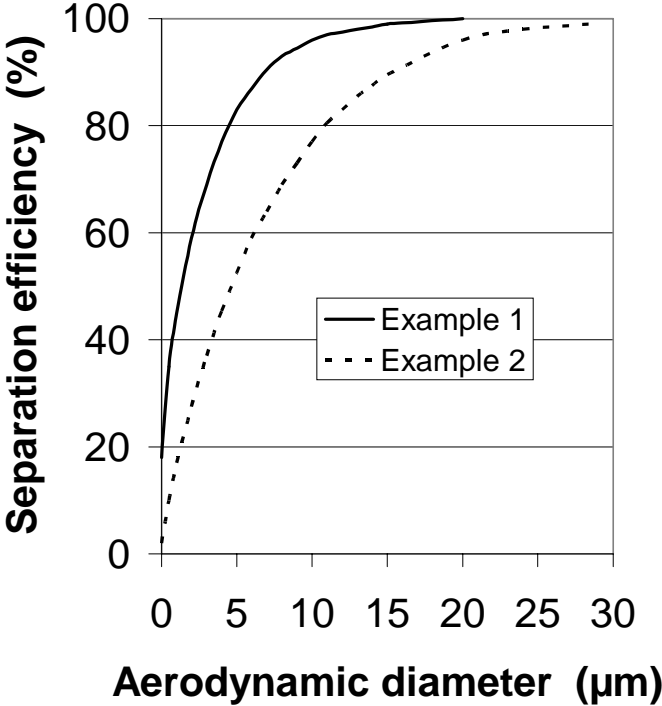


Figure 1 Typical separation characteristics of multicyclones.