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Particle emissions from biomass combustion in small combustors

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Abstract

Literature data on particle emissions are compared with emissions from combustion of wood pellets and wood briquettes in commercial small-scale combustion devices: a pellet stove, two pellet burners and two smaller district heating boilers. The influence of operating parameters and fuel quality was investigated. Mass concentration, number concentration and number size distribution of particles were determined. The mass size distribution was analysed as well as the inorganic components. Gaseous compounds were recorded to give information about the combustion conditions. The mass concentrations of particles were between 34 and 240 mg Nm⁻³, increasing during unsatisfactory operation conditions. The number concentration was in the range of 10⁷–10⁸ particles per Ncm⁻³. The particle emission was dominated by submicron particles (size < 1 μm), both from number and mass perspective. The main inorganic components of the submicron particles were potassium, sulphur, chlorine and oxygen. Small amounts of sodium, magnesium and zinc were also found. The contents of potassium, chlorine, and sulphur in the fuel are important for the composition of the emitted inorganic submicron particles.

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Keywords: Particle emissions; Biomass combustion; Domestic heating; District heating

1. Introduction

Numerous epidemiological studies have shown a correlation between negative health effects and increased concentrations of particulate matter in the ambient air (e.g. [1,2]). Because of this correlation and the increasing use of biomass combustion, the particle emission from biomass combustion, particularly that of ultrafine particles, needs further investigation. The present work is limited to primary combustion particles emitted from domestic and small district

heating boilers. There are two main sources of such particles from small-scale biomass combustion:

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

The aim of this work is to assess the properties of particle emissions from small-scale biomass-fired furnaces by summarising the literature and by comparing with examples of particle emission from combustion of the “new” fuels wood pellets and wood briquettes.

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The particle emission is discussed in terms of the following parameters:

- Mass concentration and mass size distribution.
- Number concentration and number size distribution.
- Chemical composition.

Mass concentration is mass of particles per unit volume of flue gas, while mass size distribution is mass concentration distributed over particle size. Number concentration of particles is number of particles per unit volume of flue gas, and number size distribution is number concentration distributed over particle size. The measurements performed in this work focus on small combustors with thermal outputs ranging from a few kW up to 10 MW, usually without advanced dust cleaning. To make the emission picture more complete, information on larger boilers is also included in the introduction and the discussion sections. The combustion devices are coarsely sorted up into domestic furnaces and large-scale boilers. The domestic furnaces include uncontrolled open fireplaces as well as controlled devices, such as wood-chip burners. All large-scale boilers are controlled. In the large boilers, the particle data are regarded either upstream or downstream of the dust cleaning devices.

1.1. Combustion of biomass in domestic furnaces

Mass concentrations of particles in the flue gas from domestic combustion of wood pellets, wood logs and wood chips have been reported to be in the range of 30–1500 mg Nm⁻³ [3–8]. The highest mass concentrations were recorded [3,5] during combustion of wood logs in a wood stove or a fireplace, i.e. in uncontrolled combustion devices. Muhlbaler Dasch [3] investigated the emissions of wood species and the influence of the size of the wood logs. He observed that combustion of large wood logs led to higher mass concentration of particles than split wood, and related this to unsatisfactory combustion conditions with low burning-rate, temperature and gas flow rates. The unsatisfactory combustion conditions in the residential wood fireplaces investigated by Muhlbaler Dasch [3] gave a uni-modal size distribution with a maximum at about 170 nm. This location of the maximum is similar in flue gas from automatically operated wood

furnaces, for which values between 50 and 200 nm have been reported, except for bark that has a second maximum around 5 μm [9]. Also automatically operated wood furnaces have been reported to produce two maxima in the mass size distribution: one at a particle size smaller than 400 nm and one at around 10 μm, thus showing a bi-modal mass size distribution [8].

The number concentrations of particles emitted from domestic combustion show large variations in the range of 10⁷–10¹⁰ particles per Ncm³ [6–8,10,11]. The level of excess air seems to affect the number concentration and is also an indicator of combustion conditions. For example, the number concentration of particles emitted from combustion in automatically operated wood chip burners decreases in most cases when the oxygen concentration in the flue gas is reduced [6–8,10]. During batch-wise firing of wood logs the combustion conditions clearly vary between the different combustion phases. Hence, in a batch-fired wood stove the maximum in the number size distribution was found at the largest particle size during the start-up phase, at a lower size in the intermediate phase and at the lowest during burn-out [6,7,10]. The number size distribution typically peaks around 200 nm during start-up, at 150 nm during the intermediate phase, and at 50 nm during the burn-out phase. A displacement of the maximum in number size distribution towards larger particle sizes when the oxygen concentration decreases is also observed for particle emissions from automatically operated wood chip burners [6–8,10]. The maximum in the number size distribution then varies between 60 and 100 nm [6–8,10] (in one case for wood chips this trend was not consistent through the entire measurement range [10]). Besides the examples of wood stoves and automatically operated wood chip burners a submicron maximum in the number size distribution has also been observed at around 80 nm in the flue gas from an automatically operated wood pellet boiler [7] and between 70 and 90 nm for domestic wood boilers [7,12].

The chemical composition of the particles emitted during domestic combustion is closely connected to the combustion conditions, and thus to the concentrations and size distributions. High mass concentrations were dominated by organic material [3]. Rau [13] showed that unsatisfactory combustion conditions

give particles mostly consisting of organic material, and that particularly under sub-stoichiometric conditions and at low temperatures the particles from a wood stove mainly consist of organic carbon. However, at higher temperatures, the aerosol composition contains less carbon and larger fractions of potassium, chlorine and sulphur. In a paper that summarises results from different investigations Nussbaumer and Hasler [9] state that well designed, automatically operated wood furnaces may have a carbon concentration in the fly ash of below 1–10%.

1.2. Large-scale combustion of biomass

Mass concentrations of particles downstream of the convective pass (upstream of the dust separation devices) for different biomass fuels and boilers vary between 60 and 2100 mg Nm⁻³ [14–18]. These mass concentrations are reduced by dust-separation devices to appropriate levels before the flue gas is emitted to the ambient air. Just as for the mass concentrations, the following summarised data concern conditions upstream of the dust separation devices. Mass size distributions in emissions from small district heating plants are uni-modal with a maximum around 100–200 nm in grate-fired boilers [18–20] and 400 nm in boilers [17] firing straw. However, for circulating fluidised bed boilers bi-modal mass size distributions containing one submicron and one supermicron maximum have been observed [14–16]. These distributions were dominated by supermicron particles, and fine particles were only a minor part of the particle mass downstream of the convective pass. More knowledge is needed to state with certainty that the data quoted are characteristic for the type of combustors mentioned.

During combustion of straw, the concentration of submicron particles has been reported to 10⁷–2 × 10⁷ particles per Ncm³ and the maximum in the number size distribution to 150–300 nm. This is close to the results from small grate-fired furnaces, where maxima in the number size distribution were around 100 nm [21–23]. Kaufmann [23] observed a tendency to displacement of the maximum in number size distribution to larger particle sizes when the oxygen concentration decreases, in agreement with the results from domestic combustors.

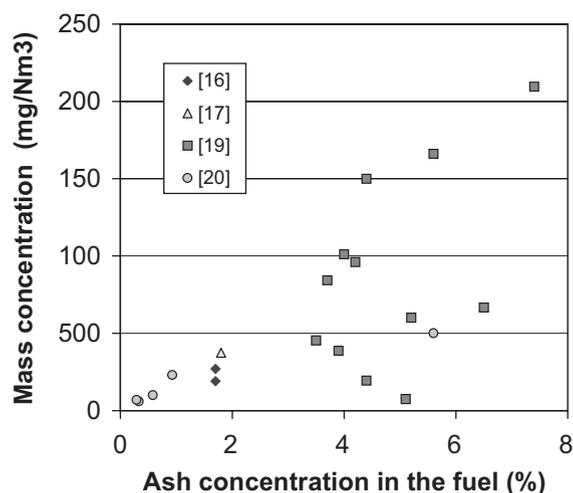


Fig. 1. Mass concentration in flue gas downstream of the convection pass (of large boilers) plotted versus ash concentration in the fuel.

Large boilers are generally operated at an appropriate oxygen concentration, and consequently under favourable combustion conditions. Particles originating from incomplete combustion are few or none, and the organic content of the particles is negligible [15,17,19]. Hence, the combustion particles originate from complete combustion. Then, the submicron particles are mainly composed of the alkali salts potassium chloride and potassium sulphate [15–17,19,20,24], while supermicron particles contain refractory species such as calcium, magnesium, silicon, phosphorous, and aluminium [15,16,24]. The chemical composition of the combustion particles shows that the particles originate from the fuel ash. The mass concentration of particles in the flue gas downstream of the convection pass (Fig. 1) even seems to correlate with the ash concentration in the fuel: increased potassium, chlorine, and sulphur concentrations in the fuel lead to increased mass concentrations of particles in the flue gas [25]. These trends suggest that the fuel composition is important for the composition of the particles emitted.

1.3. Formation mechanisms for ash particles during biomass combustion

Laboratory studies of pyrolysis of wheat straw show that the alkali metals are released in two characteristic

temperature intervals [26]: a small fraction leaves the fuel below 500°C, whereas the major part is released above 500°C. At higher temperatures, the alkali metal release is correlated with the chlorine content. A study of chlorine emissions during combustion of herbs and wood [23] shows that fly ash was mainly formed from evaporation of inorganic compounds, such as KCl, which may undergo subsequent chemical transformation, for example to K_2SO_4 in the presence of SO_2 . Furthermore, Kaufmann [23] suggested that HCl primarily forms during the devolatilisation stage but also as a by-product of the formation of K_2SO_4 from KCl. The initial feedstock composition was the most important factor for the amount and species of alkali metals released during combustion of switchgrass [27]. The switch grass contains high amounts of both potassium and chlorine, and the dominant alkali species released was potassium chloride. Combustion temperature, oxygen concentration of the emitted flue gas, and moisture content in the fuel seem to have small influence on the alkali release [27]. Four mechanisms of alkali release during combustion of biomass were identified by Dayton and Milne [28]:

- (1) For woody fuel with relatively low alkali and chlorine contents, alkali is primarily released through vaporisation or decomposition of alkali sulphate.
- (2) During combustion of fuel with high concentration of alkali and chlorine, for example herbaceous fuel, grass, and straw, alkali 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 leads to release of alkali cyanide as the dominant compound.

Dayton et al. [27] showed that adding steam to the combustion environment shifted the release of alkali from alkali chloride to alkali hydroxide. Thus, this study indicates an influence of fuel moisture [29].

For combustion of straw, Christensen [17] 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 mechanisms for formation of aerosol during straw combustion:

- (1) KCl is partially converted to K_2SO_4 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 SO_3 .
- (3) Nucleation of K_2CO_3 or K_2SO_3 takes place prior to the condensation of KCl and K_2SO_4 . The potassium condenses as K_2CO_3 , K_2SO_3 and KCl. K_2SO_3 and KCl react to form K_2SO_4 .
- (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 K_2SO_4 may condense on these particles at lower temperatures as in mechanisms 1 and 2.

Obenberger et al. [18] identified processes of aerosol formation for wood chips and bark. The mechanisms were developed from field measurements and characterisations of combustion particles in grate furnaces. The principal proposals are:

(1) *Wood chips*: Potassium sulphate and chloride are formed by homogenous gas phase reactions and then nucleate homogeneously and/or condense on existing surfaces. Calcium is a part of the coarse particle fraction, but it interacts with the submicron fraction by offering surfaces for condensation of metal vapours.

(2) *Bark*: Very small calcium particles exist in the flue gas leaving the fuel bed, because thermodynamic data show that release of calcium to the gas phase is not possible. Potassium chloride and potassium sulphate probably condense on the surface of the calcium particles, but they also nucleate homogeneously. Supermicron particles contain increasing amounts of calcium and magnesium. They are entrained from the fuel bed by the flue gas.

2. Materials and methods

Additional particle measurements on small-scale biomass combustion were carried out in the present

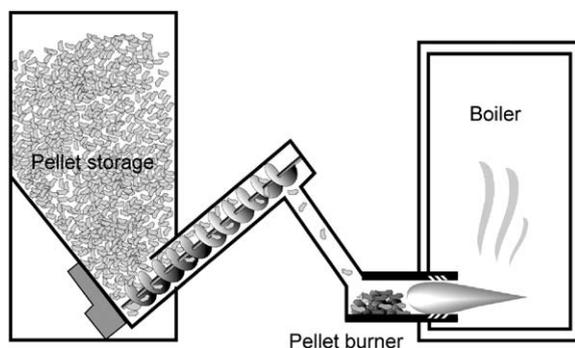


Fig. 2. Schematic picture of a domestic pellet burner.

work. The materials and methods used during these measurements, as well as the measurement set-up, are briefly presented here. Details are given in Johansson [25].

2.1. Combustion devices and measurements

Particle emissions were recorded from two size classes of boilers: small boilers for domestic heating and slightly larger boilers for small district-heating systems.

Domestic heating: Two domestic pellet burners and one pellet stove were investigated 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, which results in a cyclic, intermittent operation of the pellet burner. One of the pellet burners has a maximal thermal output of 11 kW and is supplied with a pilot flame (a small flame that is maintained also when there is no heat demand). The other pellet burner has a maximal thermal output of 22 kW and is electrically ignited. A schematic picture of a domestic pellet burner is shown in Fig. 2. The pellet stove has a thermal maximal output of 6 kW. A picture of the stove is shown in Fig. 3. The emissions were sampled in the chimney around one metre downstream of the combustors. The pellet burners were operated continuously at maximal thermal output (optimized combustion conditions. In the pellet stove, the influence on the emission of the fuel feed rate, and consequently of the thermal output, was studied.

District heating: Two 2 MW_{th} grate-fired district heating boilers of basically the same design are

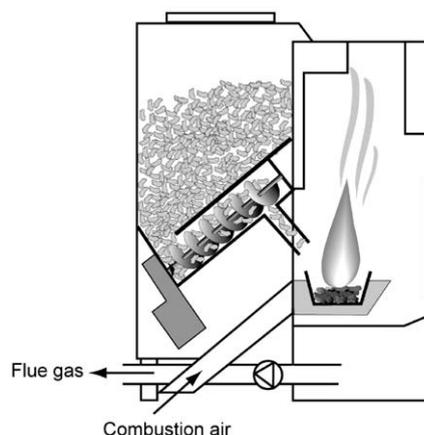


Fig. 3. Schematic picture of the pellet stove used in this work.

considered, one fired with wood pellets, and the other with wood briquettes. A schematic picture of the district heating boilers is shown in Fig. 4. The purpose was to characterise particle emissions, and therefore the measurements were performed downstream of the multicyclones. The operation parameters excess air, boiler load and density of fuels were varied. In the briquette boiler, in addition, a mixture of wood briquettes and dried forest residue was also run. Mean values of emissions were based on one hour or longer recordings in each operation case.

2.2. Fuels

Both the wood pellets and the wood briquettes have a cylindrical form. In the domestic pellet burners and in the stove, pellets of a diameter of 8 and 6 mm, respectively, were fired. The diameter of the pellets used in the district heating boiler was 8 mm. The wood briquettes (Table 1) have a diameter of 75 mm. The fuels were produced from sawdust and wood shavings and did not contain any additives. The chemical composition of the fuels was rather similar, as seen in Table 1. In the briquette boiler, dried forest residue was also fired. This fuel consisted of splintered and dried branches and tops of trees, which have higher ash content and considerably more sulphur and nitrogen than the pellets and the briquettes. Pellets of different densities but from the same raw

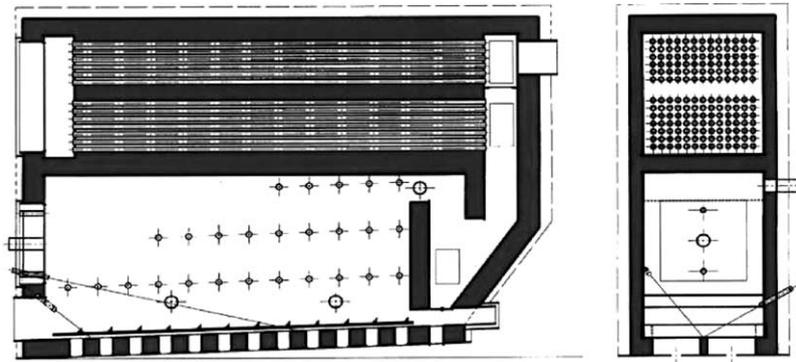


Fig. 4. Schematic picture of the district heating boilers used in this work.

material, and briquettes of normal and low density were also used. Low-density fuels disintegrate more easily and generate more fines, which affects the combustion conditions. One combustion case was run with a mixture of 75 vol% forest residue and 25 vol% briquettes.

2.3. Measurement techniques

The concentrations of CO, CO₂, O₂, NO_x and THC (total hydrocarbons) were measured continuously to relate the particle emission to the oxygen concentration and to control the combustion conditions. Particles were sampled isokinetically in two separate lines, one for total mass concentration and another for number concentration and size distribution. In the latter sampling line the flue gas was extracted and diluted in two steps. The first dilution took place under heated conditions to avoid condensation and particle growth. Number particle concentration and size distribution were measured with an (electrical low-pressure impactor) (ELPI) and mass size distribution using a (Dekati low-pressure impactor (DLPI), manufactured by Dekati Ltd). Both instruments measured concentrations and sizes in the range of 30 nm to 10 μm. The total mass of particulate emission, often called dust, was analysed gravimetrically. Qualitative analysis of the elemental composition of the submicron particles was performed by energy dispersive X-ray analysis (EDX) and the compounds were determined by time-of-flight secondary ion mass spectrometry (TOF-SIMS). As alkali compounds are

known to be important constituents of the submicron particles, a new method developed by Sjövall et al. [30] was used to investigate the fractions of potassium sulphate, potassium chloride, potassium carbonate, and the corresponding compounds of sodium.

3. Results and discussion

The mass concentration of particles from combustion of wood pellets and wood briquettes was between 34 and 240 mg Nm⁻³. The highest concentration occurred during combustion of wood briquettes at high excess air in the briquette district-heating boiler, probably caused by bottom ash particles entrained by the high-velocity gas flow. Another consequence of the high gas flow may be unsatisfactory function of the multicyclone.

In a few cases the mass size distribution was determined. The particle mass emission was dominated by submicron particles with a maximum size in the range of 80–200 nm. In the briquette boiler a tendency to 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.

Number concentrations in the emissions from the district-heating boilers varied between 0.5×10^7 and 6.5×10^7 particles per Ncm³, while those from the domestic heating devices varied between 1.4×10^7 and 13.4×10^7 particles per Ncm³. The maxima of the number size distributions were between 80 and

Table 1
Elemental analyses, heating values, and some inorganic components of the fuels

	District heating, reference pellets	Pellet burner, wood pellets	Pellet stove, wood pellets	District heating, reference briquettes	District heating, forest residue
Ash	0.3	0.5	0.4	0.3	3.2
Moisture	7.7	7.6	7.1	8.8	15.8
Lower heating value (MJ/kg)	19.12	18.98	19.01	18.80	19.20
<i>Content</i>					
Carbon	50.6	50.2	50.4	49.9	50.8
Oxygen	43.1	43.2	43.0	43.4	39.4
Hydrogen	6.0	5.9	6.0	6.2	6.0
Nitrogen	0.08	0.08	0.11	0.09	0.66
Calcium	0.07	0.10	—	0.06	0.49
Potassium	0.03	0.04	—	0.04	0.20
Sulphur	< 0.01	< 0.01	< 0.01	< 0.01	0.04
Chlorine	< 0.01	—	—	< 0.01	0.02
Sodium	0.002	0.009	—	0.001	0.050

The moisture is in mass percent related to wet fuel and the other data are in mass percent related to dry fuel.

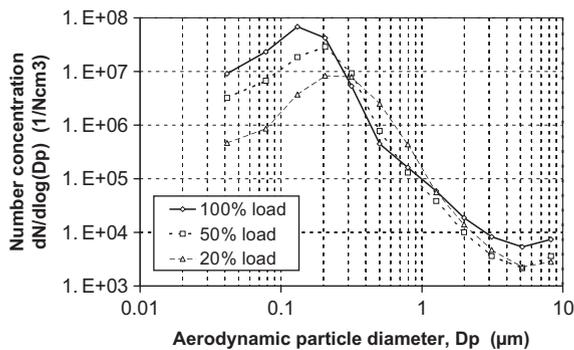


Fig. 5. Number particle size distributions at different loads of the briquette boiler (the concentrations are normalised to 13% CO₂).

300 nm, as seen in the examples of Fig. 5. Also from a number perspective, the particle emissions were dominated by submicron particles.

3.1. Influence of operating conditions and fuel quality

The principal results are presented in Figs. 5–10, where literature data are included for comparison. The literature data concern mostly controlled devices,

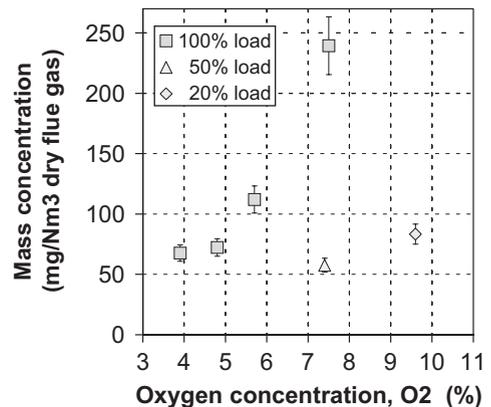


Fig. 6. Mass concentrations of particles at different boiler loads and excess air ratios during briquette combustion in the briquette boiler. The concentrations are normalised to 13% CO₂. Measurement uncertainties are illustrated by bars.

but a few uncontrolled furnaces are also included as pointed out on the figures.

Mass concentration: In the briquette boiler the four measurements at 100% load indicate that mass concentrations increase with excess air (Fig. 6). In the pellet district-heating boiler there was a slight increase in the mass concentration at decreasing load, but no clear relation between mass concentration and excess

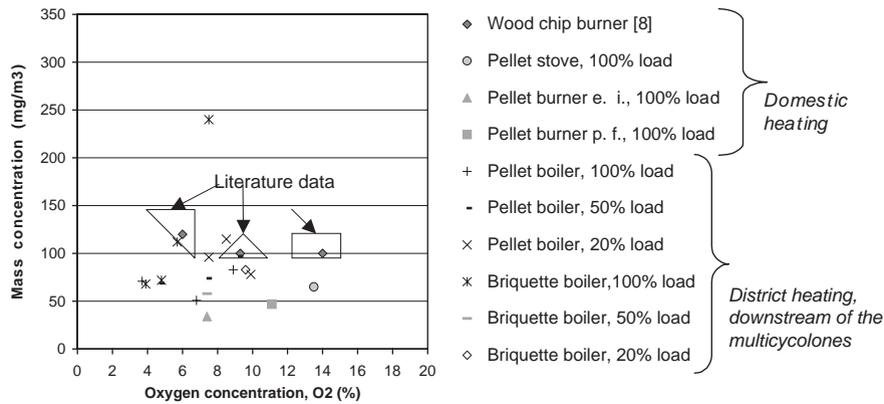


Fig. 7. Mass concentration of particles emitted versus oxygen concentration in the flue gas. The present results are shown together with literature data from controlled combustion devices.

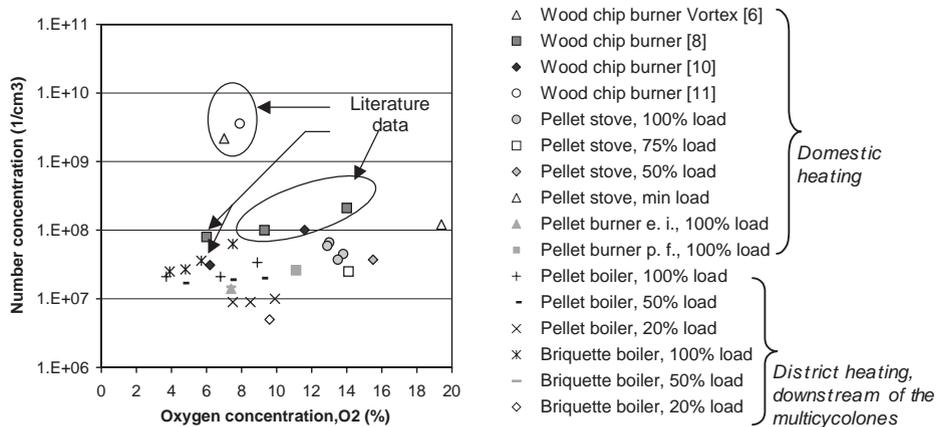


Fig. 8. Number concentration of particles emitted versus oxygen concentration in the flue gas. The present results are shown together with literature data from controlled combustion devices.

air was seen (Fig. 7). There was no obvious trend in the influence of fuel moisture on mass concentration of particles, as shown in Fig. 9.

Number concentration: In Fig. 8 the present data on number concentration are compared with literature data from domestic combustion and plotted against oxygen concentration. No clear trend is seen in the influence of oxygen on number concentration. Also, no relationship between moisture content in the fuel and oxygen concentration could be found (Fig. 10).

Number size distribution: The maximum of the number size distribution tends to shift towards larger particle sizes at low excess air and also at low load,

as seen in Fig. 5. These results are in agreement with other observations [6–8,23]. However, in one investigation [10] it was stated that reduction of the air supply as well as very high excess air ratio lead to an increase in the particle size and in the total number concentration. The differences between the results are most likely caused by burner or boiler design, and the interval of excess air levels is also important. At low excess air the number of soot particles increases, whereas at high excess air, elutriation of larger particles becomes important. Pellets and briquettes showed different influence of fuel density on the number size distribution. Combustion of low-density briquettes

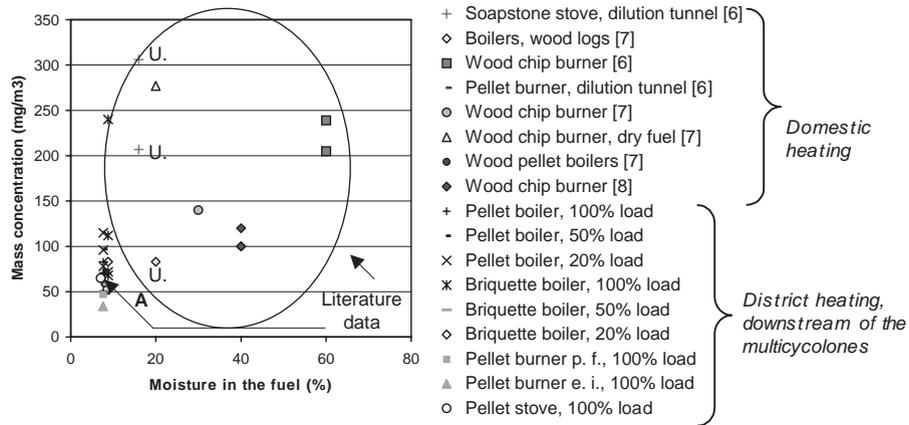


Fig. 9. Mass concentration of particles emitted versus moisture content in the fuel. The present results are shown together with literature data. The arrow labelled A indicates a single literature data point coinciding with the present value, wood pellet boilers [7]. The data are from controlled combustion devices except for those marked with U, which indicates uncontrolled combustion.

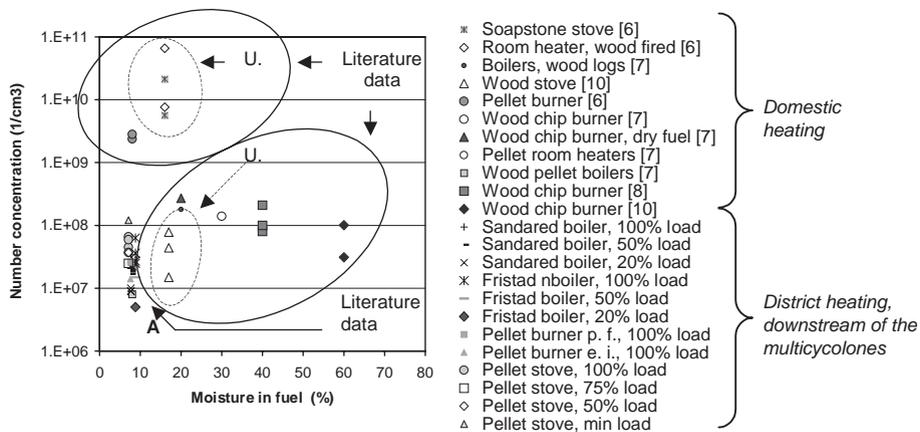


Fig. 10. Number concentration of particles emitted versus moisture content in fuel. The present results are shown together with literature data. The arrow labelled A indicates two single literature data points coinciding with the present value, pellet room heaters and wood pellet boilers [7]. The data are from controlled combustion devices except for those marked with U, which indicates uncontrolled combustion.

displaced the number distribution towards larger particle size than that of the reference briquettes. Low-density fuel disintegrates easier and may be compared with a fuel containing larger amounts of fines which causes part of the combustion to be displaced downstream of the fuel bed, and the total dust emission may increase, as well as the emission of unburnt. On the other hand, there was no significant displacement of the number size distribution during combustion of wood pellets of

different densities. In agreement with this, Oser et al. [8] reported that an increased amount of fines in the fuel results in an insignificant increase of the mass concentration of particles in the combustion gas. The reason for the divergent results may be that the two briquette qualities were more different from each other than the difference between the wood pellets and the fuels fired by Oser et al. [8].

Lower load, lower excess air ratio or low-quality fuel often mean a larger degree of unsatisfactory

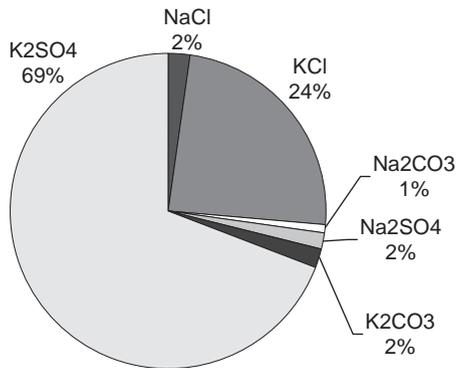


Fig. 11. Mass fractions of alkali compounds in a particle fraction collected on an impactor plate with a geometric mean diameter of 130 nm from combustion of wood pellets in the pellet burner with electric ignition.

combustion, yielding more organic condensation nuclei and more condensable gas. Such conditions favour nucleation and surface growth by condensation. In addition, low load and small air excess are typically related to lower gas flow and longer residence time. Long residence time favours particle growth by coagulation. Hence, the emitted particle size distributions are the results of a combination of several mechanisms.

3.2. Analyses of particle composition

Inorganic chemical analyses were performed on some fractions of the submicron particles. According to the EDX results, the main elements were potassium, sulphur, chlorine and oxygen. The samples also contained small amounts of sodium, magnesium and zinc. Using TOF-SIMS, the dominant alkali compound was found to be potassium sulphate, and potassium chloride was the second most important compound (in single samples potassium carbonate may, however, be more frequent than potassium chloride). An example of these results, expressed as relative mass fractions of the analysed alkali compounds, is presented in Fig. 11. The observation that potassium sulphate is the main compound in the emitted submicron particles deviates somewhat from earlier results, which claimed that potassium chloride was the main compound with potassium sulphate only second in importance [14,16,17,19,20,24]. Previous studies

of chemical composition of particles from biomass combustion focus on forest residue, hay and straw. For woody fuel, which has relatively low alkali and chlorine contents, Dayton and Milne [28] proposed that alkali compounds are primarily released through vaporisation or decomposition of alkali sulphate. For combustion of fuel with high concentration of alkali and chlorine, for example herbaceous fuel, grass, and straw, Dayton and Milne [28], on the other hand, suggested that alkali chlorides are the primary species released. The currently investigated fuels have lower concentrations of alkali, chlorine and sulphur than those quoted above, and that may explain the predominance of sulphate instead of chloride. However, it is clear that potassium, chlorine, and sulphur in the fuel are most important for the composition of emitted submicron particles. More knowledge about the mechanisms of alkali release and the subsequent formation of particles is needed, though.

4. Conclusions

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

- Combustion of biomass in domestic boilers, not supplied with particle separation, leads to particle size distributions dominated by submicron particles with uni-modal size distribution, both in the case of mass and number concentrations. This is shown for combustion of wood pellets in this study and it confirms earlier work. Also in boilers with multi-cyclones, mass and number concentrations of particles downstream of the separator are dominated by submicron particles. The mass concentration of particles increases under unsatisfactory combustion conditions due to particles originating from incomplete combustion. These include soot and condensable organic particles. Such unsatisfactory combustion conditions can result from high moisture content in the fuel, low excess air or imperfections in the boiler design.
- The excess air affects the number size distribution. In the cases investigated, the number maximum was found at larger particle sizes for low excess air than for high excess air. Low load and low-density fuel also lead to displacement of the number

mode towards larger particle sizes compared to normal-density fuel (briquettes).

- Literature data and the present study show that the contents of potassium, chlorine, and sulphur in the fuel influence the composition of the emitted inorganic submicron particles to a large extent.
- The data from different sources are quite scattered. More work is therefore needed to sort out the differences and the mechanisms of formation of particles from different biofuels.

Acknowledgements

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