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COMBUSTION AEROSOLS FROM FULL-SCALE SUSPENSION-FIRING OF WOOD PELLETS

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ABSTRACT

The objectives of the present work were to investigate the aerosol formation mechanisms during full-scale suspension firing of wood, and, to evaluate the effect of coal fly ash addition on the formation of aerosols under different ash load conditions. Tests with suspension firing of 100 % wood pellets, with and without injection of coal fly ash as additive, were carried out at the 800 MWth multifuel boiler at Avedøre Power Plant. An extractive sampling system consisting of a gas ejector-diluter connected to a 10-stage Berner type low pressure cascade impactor (aerodynamic diameter range of 0.03 – 12.7 μm) was used to sample aerosols in the flue gas, in the top of the boiler before the SCR ($T_{\text{fluegas}} \sim 350$ °C). The collected aerosols were subsequently characterized with respect to particle size distribution, morphology, and chemical composition. The mass-based size distribution of the aerosols revealed that the formation of submicron particles was increased significantly when no coal ash was injected, as compared to the reference experiments with addition of coal fly ash. PM_{10} for the experiments without coal ash addition was in the range 44 – 47 mg/Nm^3 , while it was only 11 – 19 mg/Nm^3 for the experiments with coal fly ash addition. This indicates that the coal fly ash is effective in capturing volatile alkalis released from the wood during combustion, thus suppressing the homogeneous nucleation of alkali-salts. SEM/EDS and TEM/EDS analysis revealed that the large condensation peak from pure wood combustion (without coal ash addition) consisted primarily of irregularly shaped aggregates rich in K, Cl and S (probably KCl and K_2SO_4). The addition of coal fly ash mainly affected the submicron aerosols in two ways: the relative amount of spherical particles originated from molten minerals was increased; and the composition of the aggregates was changed from K-Cl-S rich to Ca-P-Si rich. In conclusion, the results confirm that coal fly ash is effective in capturing gaseous K, presumably by incorporating gaseous K into solid potassium-aluminum-silicates, thereby reducing significantly the amount of submicron aerosols formed from homogeneous nucleation, coagulation and condensation of KCl and K_2SO_4 . A potential for optimizing the coal fly ash concentration exists.

Keywords: biomass suspension-firing, combustion aerosols, alkali metals

1 INTRODUCTION

Tests with suspension firing of 100 % wood pellets, with and without injection of coal fly ash as additive, were carried out at the 800 MW_{th} multifuel boiler (AVV2) at Avedøre Power Plant, near Copenhagen, Denmark. During normal operation of the boiler, coal fly ash is routinely injected as alkali-capture additive when wood pellets are used as fuel; however, the positive effects are not fully clarified. The main objective of the present campaign was to clarify in more details the specific effects of adding coal fly ash as additive during suspension-firing of wood pellets. As part of the measuring program, a low pressure cascade impactor was applied to sample the aerosols in the flue gas, and the collected aerosols were analyzed by SEM/EDS and TEM/EDS.

In the present paper, the mass-based particle size distributions, particle morphology, and chemical composition of the aerosols collected during the campaign are presented and discussed.

2 MATERIALS AND METHODS

2.1 Experimental

The full-scale test campaign was based on “commercial” wood pellets (Baltic and North American), and coal fly ash obtained from another pulverized coal-fired boiler (Asnæs Power Plant). Different concentrations of fly ash were injected, as additive, during combustion tests fired with 100 % wood pellets at full-load conditions. The amount of fly ash injected was 6 ton/h (corresponding to normal “reference” conditions), 3 ton/h, and 0 ton/h, respectively. As the load of wood pellets is ~130 – 140 ton/h at full-load conditions, and the ash content of the wood is around 1wt. %, this corresponds to a coal ash to wood ash ratio of ~4, ~2, and 0, respectively. Natural gas was used for start-up of the boiler and as supplementary fuel when needed. A schematic overview of the different tests is shown in Table 1.

Table 1: Aerosol measurements performed during the test campaign at AVV2.

<i>Date</i>	<i>Experiment name</i>	<i>Coal ash load</i>	<i>Comments</i>	<i>Number of LPI measurements</i>
3/12	WP+6t/h ash	6 t/h (reference)	Boiler start-up problems ¹	3
6/12	WP+3t/h ash	3 t/h		4
7/12	WP	0 t/h (pure wood)		4
8/12	WP+6t/h ash_2	6 t/h (reference)		3
9/12	WP_2	0 t/h (pure wood)		3

¹: Only 2 (of 3) wood mills were running at LPI measurement 1 and 2

A 10-stage Berner type low-pressure cascade impactor (LPI) with an aerodynamic diameter range of 0.03-12.7 μm was used to sample aerosols in the flue gas. A schematic drawing of the setup is shown in Figure 1. The setup has previously been applied to investigate the aerosol formations during e.g. co-firing of Solid Recovered Fuel and coal at a pulverized coal-fired boiler [1], and during grate-firing of straw with addition of sorbents [2].

During the present experiments, flue gas with a temperature around 350 °C was withdrawn by a gas ejector located before the SCR, at the top of the boiler. The probe was placed in a vertical direction at an angle of $\sim 90^\circ$ to the direction of flue gas, as indicated in Figure 1. With the present set-up, the ejector capillary, which is the inlet of the flue gas, acts as a prefilter with a cutoff diameter of approximately 3 μm . By using the ejector it is ensured that large fly ash particles are filtered from the sampling line by inertial forces while the sampling of submicron particles is not affected. In order to prevent water condensation, reduce inorganic species coagulation, and extend the sampling time, the withdrawn flue gas is diluted and quenched by dry and filtered pressurized air in the ejector probe before directed to the LPI. The dilution ratio is determined by measuring the CO_2 concentrations in the undiluted and diluted flue gas through an IR CO_2 analyzer (Rosemount, NGA 2000). In this study, the dilution ratios measured in different experiments were between 15 and 20.

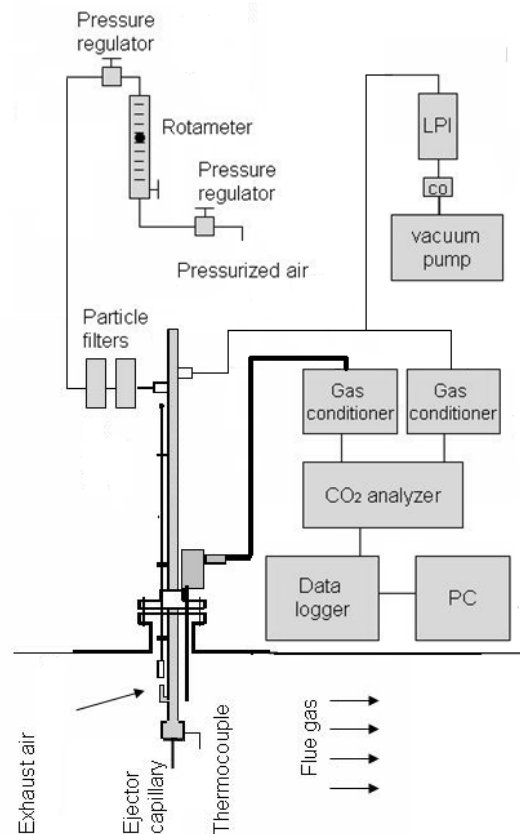


Figure 1: Ejector diluter and low-pressure cascade impactor for aerosol measurements.

The ejector was connected to the LPI by a carbonized tube which could minimize particle deposition caused by static electrical charges. In the LPI, particles were collected on aluminum foils coated with a thin film of Apiezon H grease using a dilute toluene solution of the grease. In order to avoid water condensation, the LPI was maintained at 80 °C during the experiments by a heated jacket. The cut-diameters of different stages in the impactor at this temperature were calculated by assuming constant values of the Stokes number. The experiments lasted for 30 minutes to obtain representative and sufficient

aerosol samples. Three or four measurements were performed at the same operational condition in order to confirm the reproducibility of the experiments.

2.2 Analysis methods

The Al-foils from the LPI were weighted before and after sampling with a Sartorius M5D-000V001 microbalance to find the mass-based particle size distributions of the deposited particles.

The overall morphology and bulk chemical composition of the collected aerosols was studied by means of Scanning Electron Microscopy (SEM) equipped with Energy Dispersive Spectroscopy (EDS) for elements quantification, at DTU CEN. The particles for SEM/EDS analysis were transferred to a carbon sticker and coated with carbon before placed in the SEM. In addition, high resolution Transmission Electron Microscopy (TEM) equipped with EDS was used to characterize in more details the morphology and composition of the individual aerosol particles. For the TEM/EDS analysis, the particles were transferred to a Cu-grid, and the Cu content was neglected in the EDS analysis.

3 RESULTS AND DISCUSSION

3.1 Mass-based particle size distribution

Figure 2 depicts the average mass-based particle size distributions obtained from impactor stages 1 – 8 of the different experiments. Due to the filtration effect of the ejector capillary, the samples collected on the impactor stages 9 - 10 are considered not representative for their distribution in the flue gas, thus data from these stages are excluded from the figure. The error bars depict the standard deviations within the individual measurements performed at each experimental condition. It is seen that the reproducibility of the experiments is generally very good.

The total mass-loads of aerosol particles below 2.5 μm ($\text{PM}_{2.5}$) and of submicron particles below 1.0 μm (PM_1), respectively, are listed in Table 2.

For all experiments, the mass-based particle size distributions reveal a distinct, submicron peak, with a maximum around 0.1 – 0.2 μm , see Figure 2. However, the size of this peak is significantly larger, and shifted towards larger particles, for the experiments without coal ash addition as compared to the experiments with coal ash addition, as discussed in details below:

For the experiments with injection of coal ash, the distributions are not significantly influenced by the concentration of coal ash added (3 or 6 t/h, respectively). The submicron peak is rather small and centered around 0.12 μm , with only small variations between the different experiments. This distribution is comparable to previous results obtained for coal combustion [1]. The mass-load of the submicron particles below 1 μm (PM_1) varies between 11 and 19 mg/Nm^3 for these experiments, see table 2 for details.

For the experiments without coal ash addition, on the other hand, the submicron peak is enlarged significantly, indicating increased homogeneous condensation at these conditions. PM_1 for the experiments without coal ash addition is in the range 44 – 47 mg/Nm^3 (see table 2), which corresponds to an increase in the mass-concentration of submicron particles of 2.3 to 4.2 times, as compared to the experiments with coal ash addition. The peak is in this case centered around 0.20 μm .

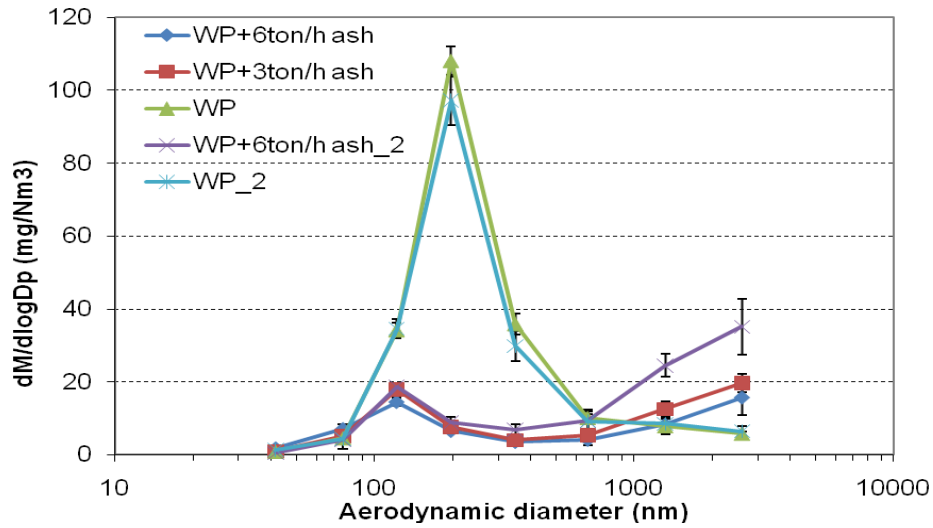


Figure 2: Average mass-based particle size distribution from different experiments. The error bars indicate the standard deviation ($n=3$ or 4 , see Table 1) between individual measurements at each experimental condition.

Table 2: Total mass-load of aerosols (PM_{1} and $PM_{2.5}$) for the different tests, as measured by LPI.

Date	Experiment name	PM_{1} (mg/Nm^3)	$PM_{2.5}$ (mg/Nm^3)
3/12	WP+6t/h ash	11.3 ± 1.0	19.4 ± 2.4
6/12	WP+3t/h ash	13.2 ± 1.3	19.8 ± 2.4
7/12	WP	46.9 ± 1.4	49.1 ± 1.3
8/12	WP+6t/h ash_2	19.1 ± 2.4	29.8 ± 2.9
9/12	WP_2	44.0 ± 3.1	45.8 ± 3.3

The mass-load values of submicron particles (PM_{1}) of $44 - 47 \text{ mg/Nm}^3$, as well as the position of the peak, for the pure wood combustion experiments, without coal ash injection, are similar to what has previously been reported for grate-firing of beech wood [3], and for wood combustion in general (softwood, hardwood and bark) [4].

3.2 Aerosol chemical compositions

Stage 2 to 8 of a selection of impactor samples was analyzed by SEM/EDS for bulk chemical composition. Representative examples of the results, depicted as chemical composition (wt. %) as function of particle size, are shown in Figures 3 – 5 for the three experimental conditions studied: 0 t/h coal fly ash, 3 t/h coal fly ash, and 6 t/h coal fly ash. It should be noted that the contents of carbon and oxygen are neglected in the SEM/EDS analyses because the method has large uncertainties in quantifying these elements and the carbon content is greatly influenced by the coating method. Thus the composition shown in Figures 3 – 5 is the relative composition of the inorganic elements. The absolute contents of carbon and oxygen are difficult to determine through SEM/EDS. Figure 3 depicts the composition of the aerosols collected from pure wood combustion. According to the figure, it is evident that the large condensation peak from pure wood

combustion consists primarily of K, Cl and S (amounting to 87 % w/w at impactor stage 3). The molar ratio $K/(Cl+2S)$ is close to unity, especially at the lower impactor stages 2 - 6 ($0.076 \mu\text{m} - 0.67 \mu\text{m}$) clearly indicating that the vaporization mode primarily is formed by condensation of potassium chloride (KCl) and potassium sulfate (K_2SO_4). However, the relatively high amounts of Cl are a little surprising, as wood usually do not contain significant amounts of Cl. It suggests that almost all the Cl present in the wood is probably released as KCl (or is reacting with K to form KCl).

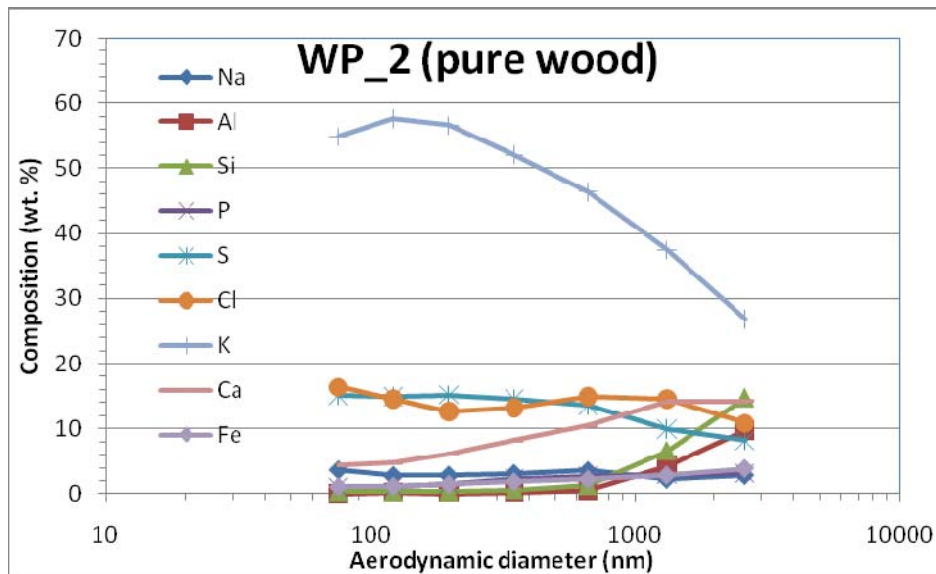


Figure 3: Elemental composition of aerosols collected during combustion test with pure wood pellets, without addition of coal fly ash.

During pure wood combustion, the content of K peaks at impactor stage 3 or 4 and then decreases continually with increasing particle size. The content of Cl and S is relatively constant (both around 15 % w/w) from stage 2 – 6 ($0.076 \mu\text{m} - 0.67 \mu\text{m}$) and then decreases slightly. The Ca content increases constantly with increasing particle size, from around 4 % w/w at stage 2 to about 14 % at stage 8. The content of Si and Al is negligible in stage 2 - 6 ($0.076 \mu\text{m} - 0.67 \mu\text{m}$), and then increases significantly with increasing particle size on stage 7 and 8 ($1.32 \mu\text{m} - 2.61 \mu\text{m}$). Similar results for concentration vs. particle size in aerosols from a pulverized wood fired boiler have been reported by Skrifvars et al. [5]. They found from impactor measurements that alkali sulphates and chlorides were enriched in the smallest sized particles, $< 1 \mu\text{m}$, while Si was found in the largest particles, $> 10 \mu\text{m}$, and Ca seemed to be enriched in the size range $1 - 10 \mu\text{m}$ [5]. Similar tendencies for concentration vs. particle size for K, Cl, S, and Ca in aerosols from grate-firing of beech have been reported by [3].

The much smaller vaporization peak (at around $0.12 \mu\text{m}$) from the experiments with addition of coal ash consists mainly of Ca (20 – 35 % (w/w)) and P (17 – 20 % (w/w)), together with a noticeable amount of K (10 – 15 % w/w) (see Figure 4 and 5). Smaller amounts of Si, Al, Fe and S are also found (generally amounting to 5 – 10 % w/w each). The content of Cl is negligible ($< 1-4 \%$ w/w) at the lower impactor stages. As compared to pure wood combustion, the significantly smaller vaporization peak and the much lower

K content (especially in the submicron particles) indicate that the K released from the wood has been captured by the added coal fly ash, probably in the form of potassium-aluminosilicates. The mechanism is probably analogy to that observed in co-combustion of coal and straw [6].

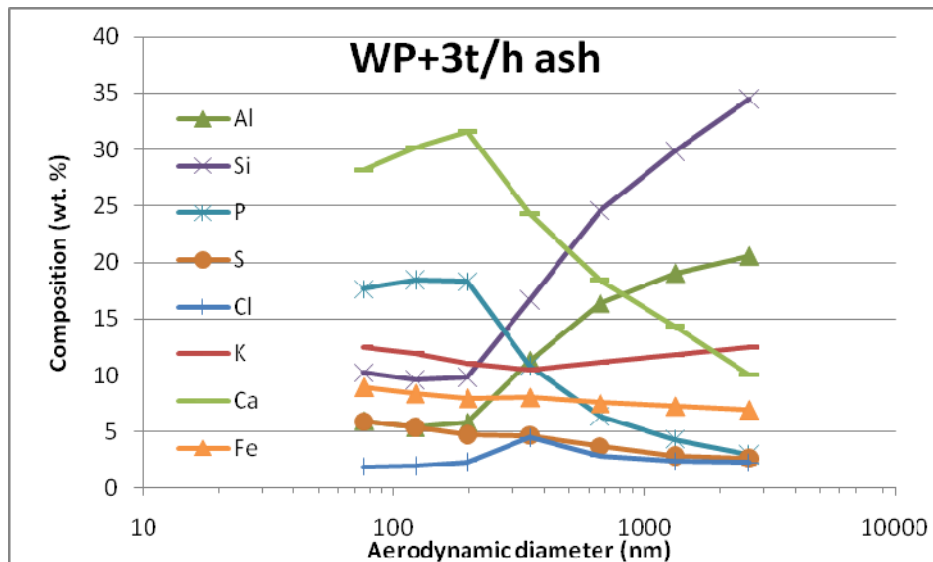


Figure 4: Elemental composition of aerosols collected during combustion test with addition of 3t/h coal fly ash

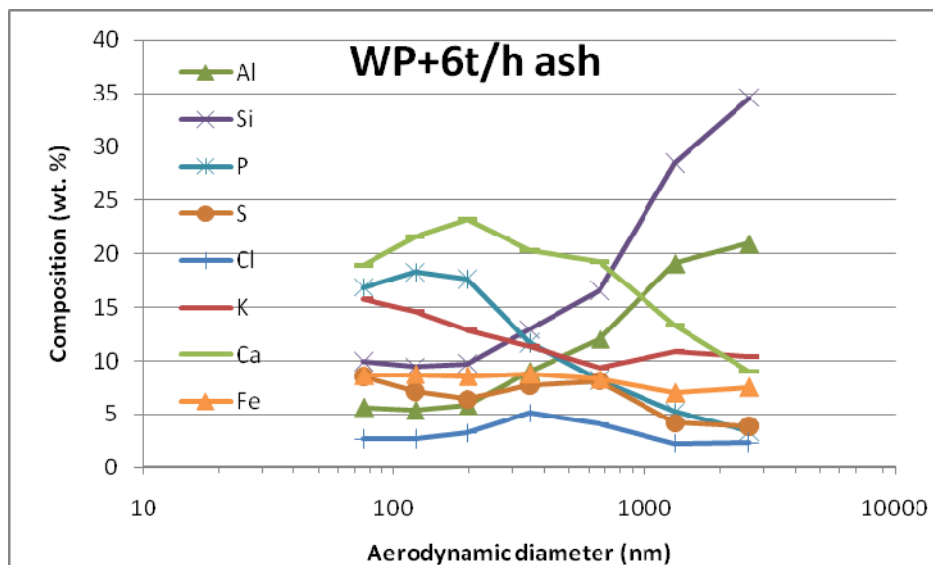


Figure 5: Elemental composition of aerosols collected during combustion test with addition of 6 t/h coal fly ash

As compared to aerosols obtained from dedicated coal combustion [1], the content of Ca, P and K in the submicron particles is significantly higher in our case, while Si and Al content is lower. This may be due to different coal composition in the two cases, or it may be a result of interactions between the added coal fly ash and the wood ash. In our

experiments as well as in the coal combustion study by Wu et al. [1], the concentration of Ca and P decreases with increasing particle size above $\sim 0.1 - 0.2 \mu\text{m}$, indicating that these elements generally have high volatilities.

3.3 Particle morphology and composition

High resolution TEM analyses reveals that the supermicron aerosols from pure wood combustion primarily consist of two types of particles: spherical particles originated from the molten minerals, and irregular shaped particles generated from the aggregation of the nucleated species. Typical examples of these two types of particles are shown in Figure 6.

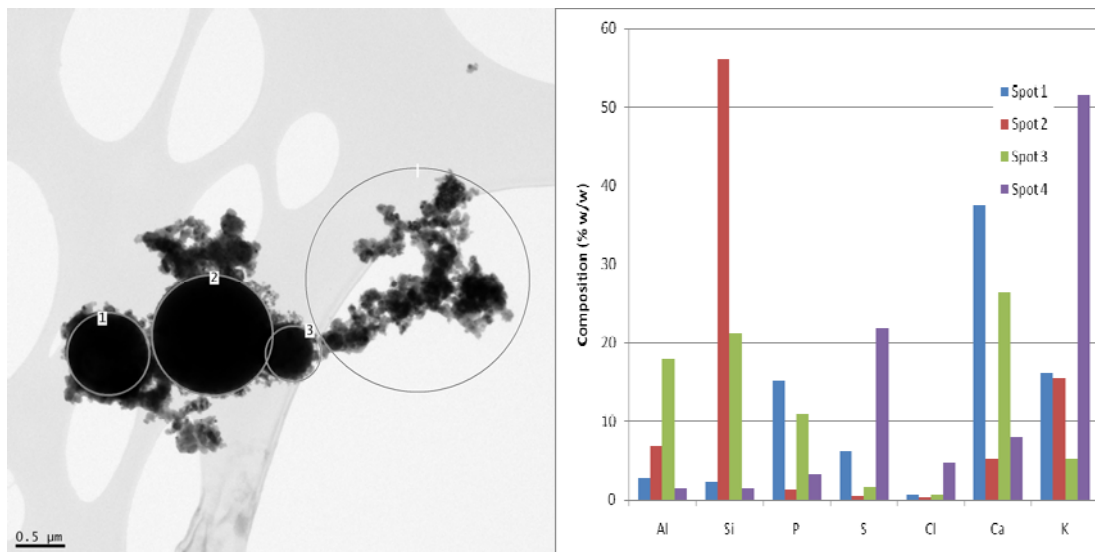


Figure 6: Typical morphology and composition of supermicron aerosols from pure wood combustion. The left picture is from TEM analysis on impactor stage 7 (mean $d_{ae} = 1.32 \mu\text{m}$); the right figure shows the corresponding spot analysis composition.

Spot composition analysis by EDS indicates that the spherical particles are mostly dominated by Si, Al, Ca, K and P (such as spot 1 – 3 in Figure 6), which is in agreement with the bulk composition analysis by SEM/EDS in Figure 3. Ca and P seem to be associated (such as spot 1 and 3). The aggregated nucleates primarily contain K and S, together with considerable amounts of Ca, Cl and P (such as spot 4). In most cases, the aggregates are found to be attached to the surface of spherical particles, but some large pieces of aggregates (with size above $1 \mu\text{m}$) are also observed.

With decreasing particle size, the contribution of the aggregated nucleates becomes more significant, on the expense of the spherical particles, as evident from Figure 6, 7 and 8.

The typical morphology of the submicron aerosols from pure wood combustion, as obtained by high resolution TEM analysis, is illustrated in Figure 7. It is seen that the ultrafine particles consist predominantly of aggregated nucleates, and that the primary particles of the aggregates usually have an irregular shape and the particle size mostly varies in the range of $20 - 100 \text{ nm}$. Compared to the vaporization mode aerosols obtained from a coal-fired power plant (Pedersen et al., 2010), the vaporization mode aerosols from wood combustion seem to have larger primary particles. Spot composition analysis of the aggregated aerosols by TEM/EDS reveals that they are primarily comprised of K, S

and Cl (Figure 8), which is in agreement with the bulk composition shown in Figure 4. However, the content of Cl tends to be slightly lower than indicated from the bulk composition. This may be a consequence of the different analysis methods.

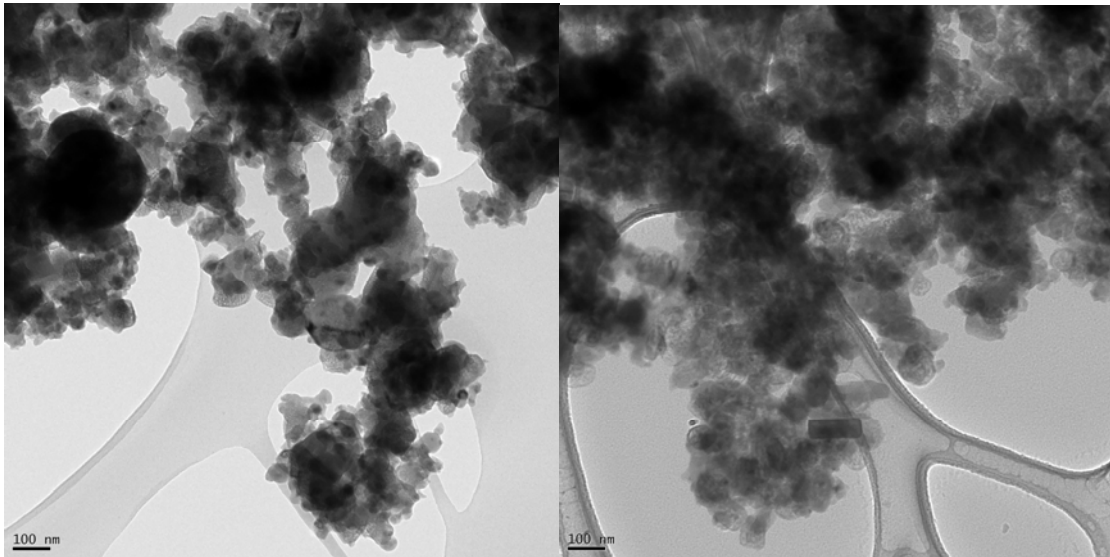


Figure 7: High resolution TEM pictures of aerosols from pure wood combustion. The left picture is from impactor stage 6 (mean $d_{ae} = 0.67 \mu\text{m}$), the right picture is from impactor stage 4 (mean $d_{ae} = 0.20 \mu\text{m}$).

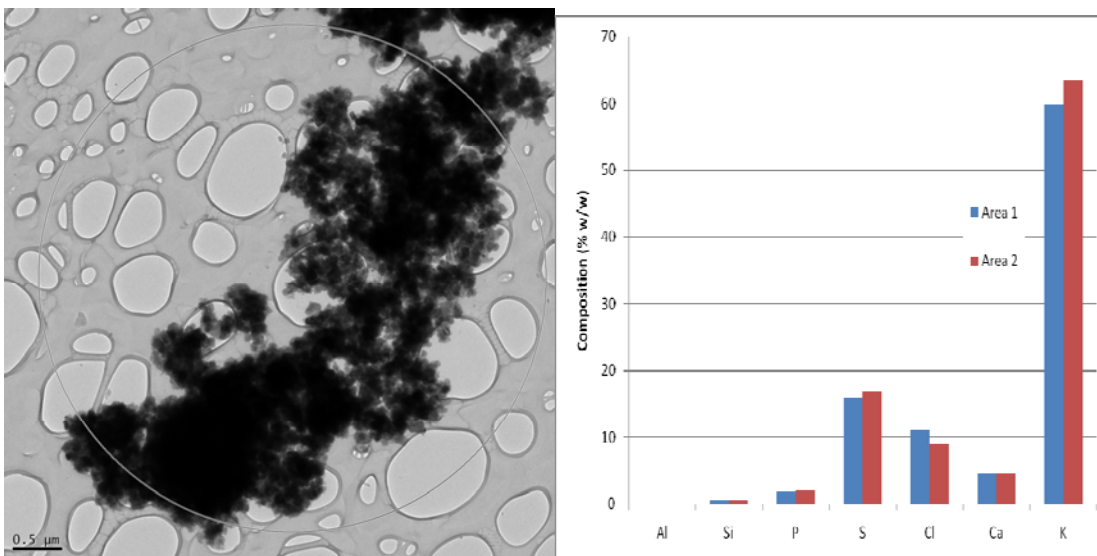


Figure 8 Spot analysis composition of ultrafine aerosols from pure wood combustion. The left figure is TEM picture from impactor stage 4 (mean $d_{ae} = 0.20 \mu\text{m}$); the right figure shows the corresponding spot analysis composition.

During the addition of coal fly ash, the collected supermicron aerosols are dominated by spherical particles, as indicated from Figure 9. EDS spot composition analysis reveals that the spherical particles generally have large K, Ca, Si and Al contents, which is consistent with the bulk compositions for supermicron particles shown in Figure 4 and 5.

Figure 10 illustrates the typical morphology of the ultrafine aerosols (mean $d_{ae} = 0.20 \mu\text{m}$) collected during the addition of 6t/h coal fly ash. It can be seen that the ultrafine aerosols mainly consists of aggregated particles. Compared to the primary particles from pure wood combustion (Figure 7), the primarily particles shown in Figure 10 seem to be smaller and more spherical.

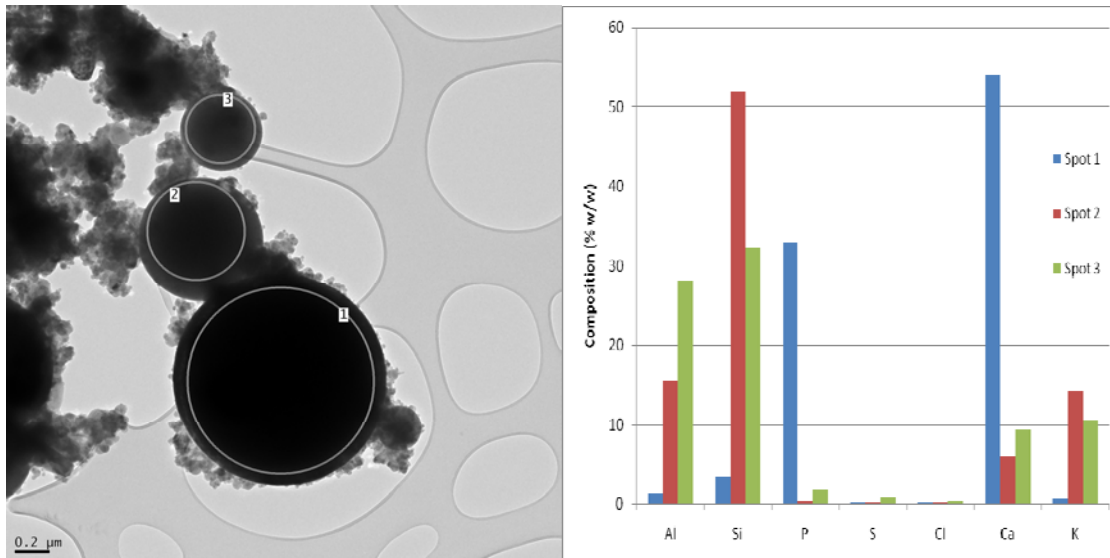


Figure 9: Typical morphology and composition of the large spherical particles from impactor stage 6 ($d_{ae} = 0.67 \mu\text{m}$) with addition of 6 t/h coal fly ash. The left picture shows the TEM picture, the corresponding spot analysis composition is shown to the right.

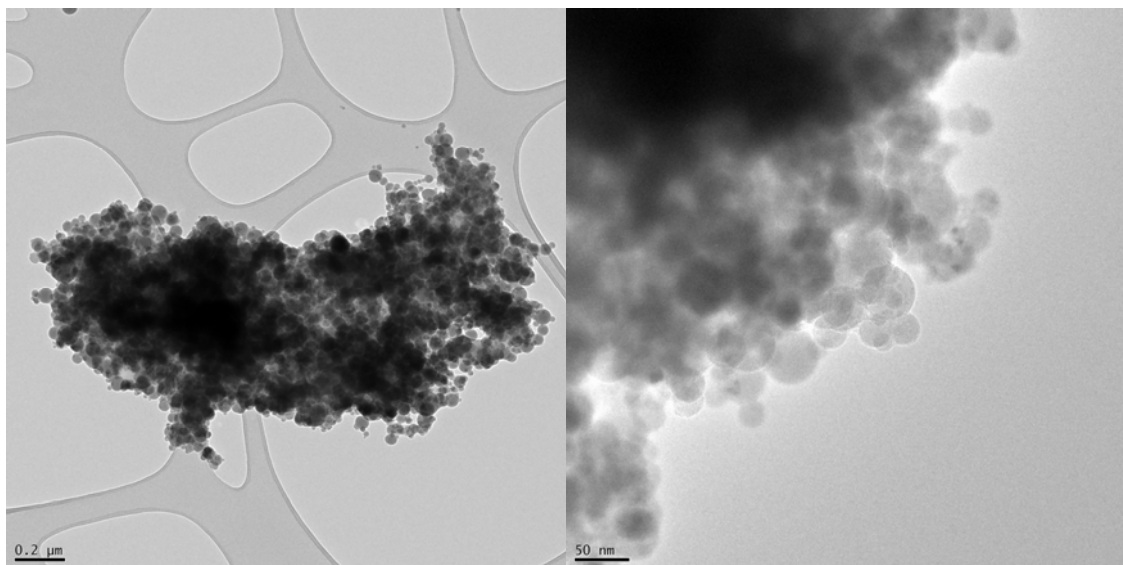


Figure 10: High resolution TEM picture of ultrafine particles (stage 4, mean $d_{ae} = 0.20 \mu\text{m}$) from addition of 6 t/h coal fly ash.

EDS analysis indicates that these particles are dominated by Ca, P and Si (Figure 11), as opposed to the K-S-Cl rich primary particles from pure wood combustion (Figure 8). It is

most likely that these particles are originated from the added coal fly ash. During pulverized coal combustion, Ca, P and Si can be vaporized under reducing conditions and form ultrafine aerosols [1]. When the coal fly ash containing these ultrafine aerosols is added to wood combustion, the aerosols may be entrained to the flue gas and result in the formation of ultrafine aerosols rich in Ca, P and Si. However, the primary particles of the ultrafine aerosols from pulverized coal combustion usually have irregular shapes [7]. The general spherical appearance of the primary particles in Figure 10 suggests that these particles may have undergone melting during flying.

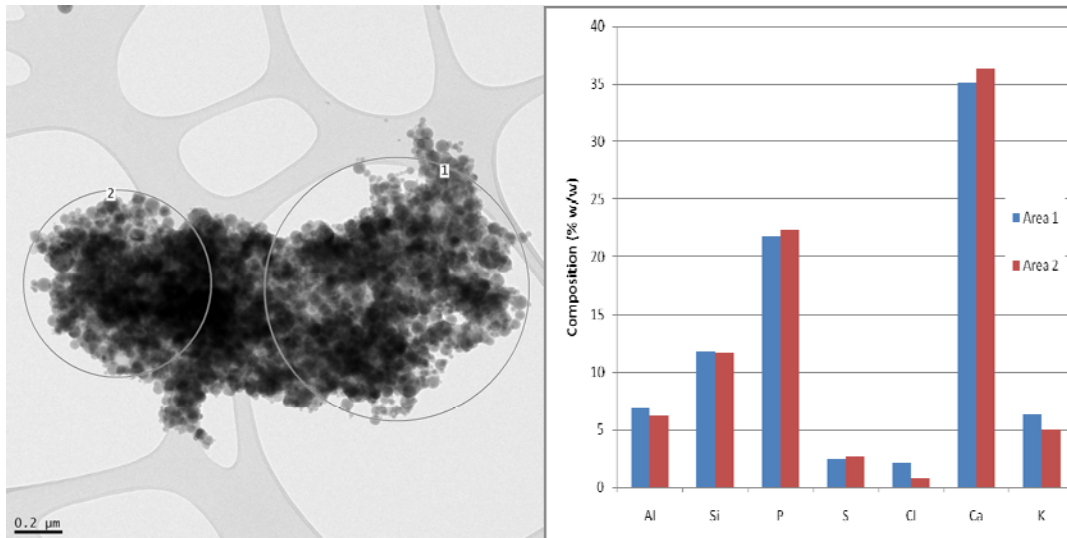


Figure 11: Spot analysis composition of ultrafine aerosols from addition of 6 t/h coal fly ash. The left figure is TEM picture from impactor stage 4 (mean $d_{ae} = 0.20 \mu\text{m}$); the right figure shows the corresponding spot analysis composition.

4 CONCLUSIONS

Full-scale aerosol measurement from tests with suspension firing of wood pellets at AVV2 revealed that the formation of submicron particles was increased significantly (2.3 – 4.2 times increase in PM_{10}) when no coal fly ash was injected, as compared to the reference experiments with addition of coal fly ash.

Analysis of the bulk chemical composition of the aerosols by means of SEM/EDS revealed that the large condensation peak from pure wood combustion (without coal ash addition) consisted primarily of potassium chloride and potassium sulphate, while for the tests with addition of coal fly ash, the submicron particles consisted primarily of Ca, P and Si. These results clearly indicate that the coal fly ash is effective in capturing volatile alkalis, and the effect is remarkable.

As there was no significant difference between experiments with addition of 3 ton/h ash or 6 ton/h ash, respectively, there seems to be a “threshold-value” for the minimum concentration of coal ash needed, which is lower than the investigated concentrations. This finding may be of interest with respect to future possibilities for optimizing the coal ash addition.

The morphology and composition of the individual aerosol particles were studied by high-resolution TEM/EDS. It was found that the submicron aerosols from pure wood combustion were dominated by irregularly shaped aggregates rich in K, Cl and S. The addition of coal ash mainly affected the submicron aerosols in two ways: the amount of spherical particles originated from molten minerals was increased in submicron aerosols; and the composition of the aggregates was changed from K-Cl-S rich to Ca-P-Si rich.

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