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PARTICULATE EMISSION REDUCTION IN SMALL-SCALE BIOMASS COMBUSTION PLANTS BY A CONDENSING HEAT EXCHANGER

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Abstract

Utilisation of biomass fuels for energy purposes has gained increasing importance as a method to reduce greenhouse gas emissions. Compared to gaseous and liquid fossil fuels the emissions of particulate matter are higher, leading to concerns about the availability of cost effective techniques to reduce aerosol emissions in small scale biomass combustion plants. In this work the applicability of reducing aerosol emissions by stimulating condensation of aerosol forming vapours on the heat exchanger furnaces is investigated. A first order estimation indicates that the heat exchanger passage has to be in the order of millimeters to obtain a higher wall condensation rate compared to heterogeneous condensation, a result verified with purpose build heat exchangers connected to a grate furnace. The measurements show that heat exchangers with an equivalent tube diameter of approximately 2 mm can reduce the aerosol emissions by approximately 70% compared to conventional boiler designs.

1 Introduction

Combustion of solid biomass offers high short-term potential for significant CO₂-reduction in energy production. Comprehensive R&D work carried out during the last decades has already led to a state-of-the-art which allows a highly efficient combustion of a wide range of solid biomass fuels at high plant availabilities as well as low gaseous emissions (Faaij 2006). However, several problems related to the formation of fine particulates during combustion, so called aerosols, are still unsolved. These problems affect both, the plant operation itself due to the formation of deposits and ash mixtures with comparably low melting temperatures as well as particulate emissions. Appropriate dust separation devices must be applied to meet current dust emission standards, which is not a problem for medium- and largescale applications since highly efficient filter systems such as baghouse filters and electrostatic precipitators are economically affordable for these installations. For small-scale (< 100 kW) biomass combustion systems, however, no highly efficient and economically sound dust precipitators are available on the market to date, which has led to significant R&D efforts in this field. This is of special importance since many European regions are currently facing severe air quality problems regarding fine particulate matter, and small-scale biomass combustion systems have already been identified as a major emission source together with industry and traffic (Jöller et al. 2005).

The particle size distribution of coarse fly ashes at boiler outlet is typically in the range of some μm to about 200 μm with a distribution peak at about 30 to 70 μm . The exact shape of the particle size distribution curve as well as its peak depend on the fuel, the geometry of the furnace and the particle precipitation in the different furnace and boiler sections but in general the particulates are above 10 μm and easy to separate using for instance a cyclone. aerosols (< 1 μm) are formed by gas-to-particle conversion processes of ash forming vapours, which have been released from the fuel during combustion. Consequently, the concentration of aerosol forming elements in a fuel, or more precisely, the release of aerosol forming elements from the fuel during combustion, is the most relevant parameter for aerosol formation. Aerosol formation cannot be influenced to a relevant extent by primary measures using state-of-the-art

plant designs and process control strategies. The basic mechanisms of aerosol formation in combustion processes are generally well known from previous research work (Friedlander 1977, Christensen 1995).

The first step of aerosol formation in biomass combustion processes is always the release of aerosol forming elements from the fuel. K, Na, S, Cl as well as easily volatile heavy metals such as Zn and Pb are released from the fuel bed to the gaseous phase. These elements subsequently undergo homogeneous gas phase reactions. The most important compounds formed are K and Na chlorides and sulphates. As soon as the partial pressure of a single compound exceeds the saturation pressure, which can be due to a high formation ratio of the specific compound, due to different thermodynamic properties of a new compound formed or due to the cooling of the flue gas, gas-to-particle conversion takes place. This can happen either by homogeneous nucleation (the formation of aerosol particles) or condensation of these vapours on existing surfaces. Nucleation and condensation are always competing processes, which means that if the existing particles provide enough surface area, nucleation might be suppressed and the dominating process is then particle growth by condensation. Once particles have been formed, however, they start to coagulate due to Brownian motion, diffusion and turbulent impaction. The typical particle size distribution of the aerosol fraction measured at boiler outlet thus depends on the time-temperature profile of the biomass combustion plant.

In Obernberger et al. (2003) it was found that a significant amount of the aerosol forming vapours already condense on the heat exchanger wall. Furthermore it was concluded that the dimensions of current heat exchanger is such that wall deposition is minimised to reduce fouling. If it is possible to minimise wall condensation however, it should also be possible to maximise it in order to reduce aerosol emissions. The basic concept is that by reducing the heat cross section of the heat exchanger passage, wall condensation is stimulated over heterogeneous condensation (condensation on pre-existing nuclei) for the aerosol forming vapours in the flue gas.

2 Concept development

2.1 Heterogeneous condensation vs. wall condensation

As soon as the flue gas temperature equals the condensation temperature of aerosol forming species in a heat exchanger, condensation on pre-existing particles as well as on heat exchanger walls take place as competing processes. To make a first order estimation of the amount of vapour which condenses on the walls of a heat exchanger, it is assumed that both the nuclei in the gas and the walls of the heat exchanger are below the condensation temperature of the aerosol forming vapours. In that case the vapour molecules diffuse both to the wall and the aerosol particles. As current design are only just in the turbulent regime ($Re = 10^4$) and a reduction in dimensions to enhance wall condensation is expected, a laminar heat exchanger passage is considered.

The mass vapour flux towards the walls of the heat exchanger per unit length $j'_{\alpha,w}$ can be written as (Bird et al. 2002):

$$j'_{\alpha,w} = \frac{Sh(D_{eq})\pi\mathbb{D}M_{\alpha}}{R_u T_g} (p_{\alpha,w} - p_{\alpha,g}), \quad (1)$$

where M_{α} represents the molar mass of the aerosol forming vapour elements, R_u is the universal gas constant, T_g is the flue gas temperature, p is the partial vapour pressure, whereas \mathbb{D} represents the diffusion coefficient of the aerosol forming vapours. The subscript α refers to the aerosol forming vapour element, whereas the subscripts w and g refer to its position, respectively on the wall of the heat exchanger and relatively far away from the wall in the

bulk of the flue gas. In equation 1, $\text{Sh}(D_{eq})$ is the Sherwood number, the ratio of the length scale (in this case the equivalent tube diameter, D_{eq}) to the diffuse boundary layer thickness. Since a fully developed laminar flow is assumed, the Sherwood number is constant at 3.66 when applying a constant wall temperature.

From measurements, see Brunner et al. (2002), it is known that the number concentration near the entrance is in the order of 10^{13} m^{-3} due to formation and coagulation of ZnO particles immediately after the fuel bed. The total particle surface area in a tube per unit length A'_p is defined as:

$$A'_p = \pi d_p^2 N \frac{\pi}{4} D_{eq}^2, \quad (2)$$

where the term πd_p^2 represents the surface area of one nucleus and N represents the number of nuclei per cubic metre, whereas $\frac{\pi}{4} D_{eq}^2$ represents the volume per unit length. Therefore, the vapour flux towards the particles in the tube per unit length can be defined as:

$$j'_{\alpha,p} = \frac{N d_p \text{Sh}(d_p) \phi \pi^2 D_{eq}^2 \mathbb{D} M_\alpha}{4 R_u T_g} (p_{\alpha,p} - p_{\alpha,g}). \quad (3)$$

Equation 3 is based on diffusion of molecules to the surface of the particle in the continuum limit. A correction factor ϕ for diffusion outside the continuum regime is introduced according to Seinfeld (1986).

Inertia effects can be neglected for particles in the sub-micrometre range (Fuchs 1964), implying that the particles follow the streamlines and the net flow around the particle is zero. Therefore the Sherwood number $\text{Sh}(d_p)$ as mentioned in equation 3 is equal to 2. A correction has to be made however for the release of latent heat during the condensation process according to Hinds (1982)

$$T_p - T_g = \frac{\mathbb{D} M_\alpha \mathcal{L} \phi}{R_u k_g} \left(\frac{p_{\alpha,p}}{T_p} - \frac{p_{\alpha,g}}{T_g} \right), \quad (4)$$

where k_g is the thermal conductivity of the surrounding flue gas. Due to the temperature dependency of the saturation pressure at the particle interface $p_{\alpha,p}$ and the particle temperature itself T_p , equation 4 cannot be solved explicitly.

The difference between particle temperature and flue gas temperature is dependent on the flue gas temperature itself and the degree of saturation $S = p_{\alpha,g}/p_{\alpha,sat}$. During the growth process, the effect of the rising particle temperature results in an increase of the partial vapour pressure near the surface of the particle, which results in a slowing down of the net condensational growth rate. Because of this the temperature difference between the particle temperature and the surrounding flue gas is relatively small for a wide range of saturation ratios.

If wall condensation and heterogeneous condensation take place at the same moment and all other particle deposition mechanisms are neglected, the absolute wall condensation rate $\varepsilon_{abs,theo}$ can be expressed as the ratio of the vapour flux towards the walls compared to the total vapour flux per unit length $\varepsilon_{abs,theo} = j'_{\alpha,w}/(j'_{\alpha,w} + j'_{\alpha,p})$. Introducing the wall saturation ratio $S_w = p_{\alpha,w}/p_{\alpha,sat}$ we can write with equations 1 and 3

$$\varepsilon_{abs,theo} = \frac{\text{Sh}(D_{eq})(S_w - S)}{\text{Sh}(D_{eq})(S_w - S) + \frac{\pi}{4} D_{eq}^2 \phi \text{Sh}(d_p) N d_p (1 - S)}. \quad (5)$$

In water cooled heat exchangers the wall temperature is by far lower than the flue gas temperature and consequently the wall saturation rate approaches zero. Assuming $S_w = 0$, figure 1 gives the result for various saturation ratios S . Decreasing the tube diameter down to typically 2 mm results in a significant increase of the wall condensation rate for all saturation ratios S to almost 90 %. Although heat exchangers with corresponding dimensions exist and are

commonly used in for instance economisers and balanced ventilation units, biomass combustion plants typically use larger heat exchanger passages (in the order of 40 mm). Therefore an intermediate experiment was performed to assess the applicability of a laminar heat exchanger in biomass combustion.

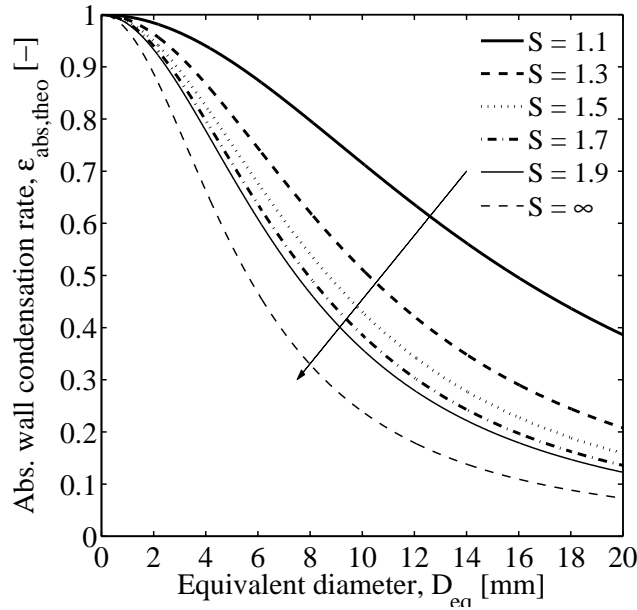


Figure 1: The effect the tube diameter D_{eq} on the absolute wall condensation rate, $\varepsilon_{abs,theo}$ for various saturation ratios. Initial conditions: $N = 1 \cdot 10^{13}$, $d_p = 0.1 \mu m$ and $S_w = 0$.

2.2 Experimental concept verification

For the experimental verification a compact laminar counterflow gas to gas heat exchanger with an equivalent tube diameter of 2.22 mm, originally developed for recuperative burners (Kemnade 2006) was used. The flue gas inlet of the heat exchanger is connected with the secondary combustion zone of the pilot-scale furnace also used in the Bioaerosol project (figure ??). The first and second ducts of the flame tube boiler are equipped with respectively 30 and 16 tubes. Furnace temperatures are measured continuously in several zones and are controlled by flue gas recirculation below and above the grate. The nominal boiler capacity amounts to 180 kW_{th}. Devices for measuring the flue gas temperature, velocity, oxygen content and the particulate emissions are installed after both the conventional and the test heat exchanger. Nine stage Berner-type low pressure impactors (BLPI) and upstream nozzles for iso-kinetic sampling are used for measuring the particulate emissions. Chemically untreated wood chips with a typical dimension of a couple of centimetres were chosen as a fuel for all experiments. With the coupled set of impactor measurements performed downstream the heat exchanger tested and the boiler it is possible to determine a PM1 emission reduction. The average relative particulate emission reductions related to the ordinary boiler design $\overline{\varepsilon_{rel,exp}}$ was calculated according to the following relation:

$$\overline{\varepsilon_{rel,exp}} = \frac{1}{n} \sum_{i=1}^n \left(1 - \frac{PM1_{test,i}}{PM1_{boiler,i}} \right) \quad (6)$$

In equation 6, $PM1_{test,i}$ and $PM1_{boiler,i}$ represents the normalised particle fraction with an aerodynamic diameter $< 1 \mu m$ of one coupled set of measurements expressed in mg/Nm^3 related to dry flue gas (13 vol% O_2). The subscripts *test* and *boiler* referred to respectively the

test heat exchanger and the ordinary boiler. To make the measurement results comparable to what is theoretically expected, losses of aerosols and aerosol forming vapours on the surfaces of the boiler which are expected to be in the range of 10 % have to be considered, see Kemenade (2005). The relation between the theoretical relative particulate emission reduction and the theoretical absolute particulate emission reduction is given as:

$$\varepsilon_{rel,theo} = \frac{\varepsilon_{abs,theo} - \varepsilon_{boiler}}{1 - \varepsilon_{boiler}}, \quad (7)$$

where $\varepsilon_{rel,theo}$ represents the theoretical relative particulate emission reduction. The absolute amount of aerosols and aerosol forming species which end up in the fire tube and the boiler are represented by, ε_{boiler} . From figure 2 can be observed that the measurement does not contradict the expected reduction in particulate matter. As also the resulting fouling layer was of powdery further development was deemed to be justified.

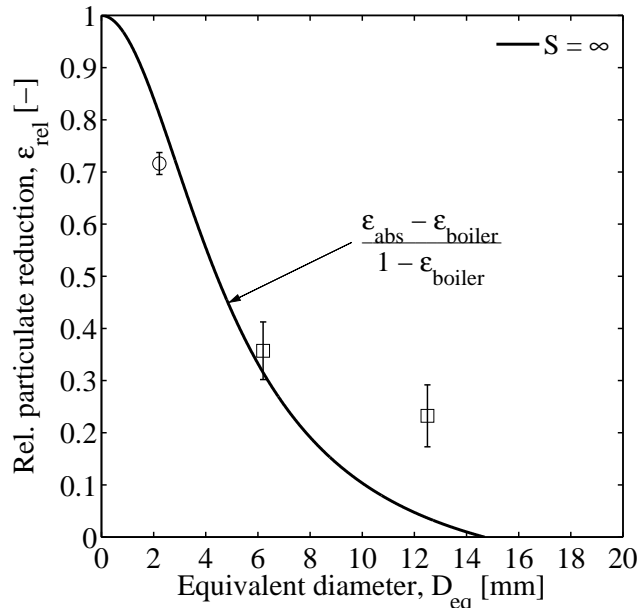


Figure 2: Theoretical (continuous and dashed line) and measured (○ = gas to gas heat exchanger, □ = water cooled heat exchanger) particulate emission reductions compared with emissions of plants with an ordinary boiler design ($\varepsilon_{boiler} = 10\%$). Measured particulate emission reductions are determined with a 95 % confidential interval.

3 Heat exchanger development

A major parameter in describing condensation is the saturation ratio, defined as $S = p_{\alpha,g}/p_{\alpha,sat}$, where $p_{\alpha,g}$ denotes the actual partial vapour pressure in the flue gas and $p_{\alpha,sat}$ the saturation vapour pressure evaluated at the flue gas pressure. A low saturation pressure is favourable to wall condensation, while increasing the saturation leads to a decrease in wall condensation and an increase in the heterogeneous condensation rate. Furthermore also homogeneous nucleation is stimulated by a high saturation ratio [?]. The work of Jensen et al. [?], describing a plug flow model for both particle formation and wall condensation is integrated in this study with the heat exchanger model. The tool used is the General Dynamic Equation (GDE) describing the change of the Particle Size Distribution (PSD) with time and and by Friedlander (1977) as

$$\frac{\partial N_k}{\partial t} + \nabla \cdot N_k v = \nabla \cdot \mathbb{D}_p \nabla N_k + \left[\frac{\partial N_k}{\partial t} \right]_{growth} + J_{d_p^*} + \left[\frac{\partial N_k}{\partial t} \right]_{coag} - \nabla \cdot v_p N_k \quad (8)$$

$\partial N_k / \partial t$ represents the change of the number concentration of particles with size class k , whereas the diffusion coefficient \mathbb{D}_p is a function of the particle size. In contrast to heterogeneous condensation, coagulation only modifies the size distribution of an aerosol, without causes a net change in the mass concentration. The flue gas velocity is denoted by v , whereas v_p is the particle velocity resulting from an external force field e.g. thermophoretic forces or electrostatic forces. The subscripts *growth* and *coag* represents the change in number concentration for particles within size class k according to respectively heterogenous condensation and coagulation. The term J is the formation of particles having a size equal to d_p^* . In Best(2007) the GDE is applied to a laminar heat exchanger passage to study the effect of a varying heat transfer rate including both homogeneous nucleation and coagulation.

The influence of the temperature profile over the length of the heat exchanger is of major interest for the design of the heat exchanger. The profile is changed by introducing an extra heat resistance in the heat exchanger walls representing for instance an insulation layer. The effect of the reduced overall heat transfer coefficient is that the particle temperature remains higher for a longer time, suppressing condensation particles while wall condensation takes place as long as the wall temperature on the flue gas side is below the condensation temperature. The price to be paid is an elongation of the heat exchanger channel and a proportional increase of the pressure drop. Figure 3 shows that a slight reduction in the heat transfer coefficient provides a significant improvement in the efficiency for larger tube diameters.

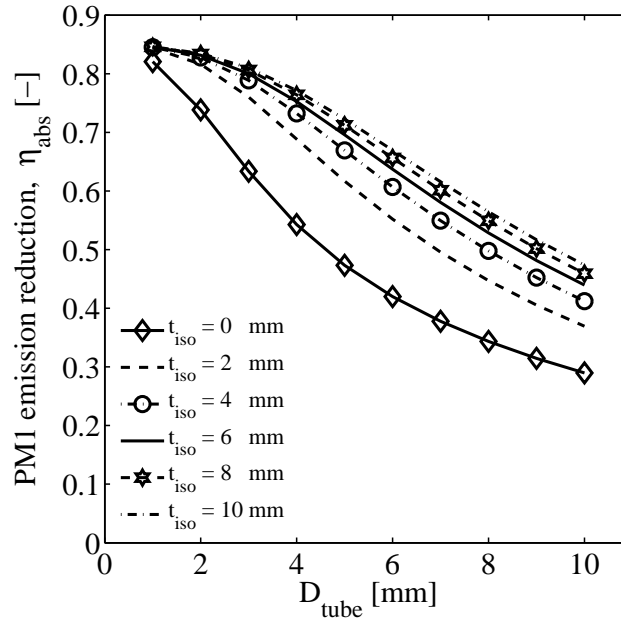


Figure 3: Effect of applying insulation material with a thermal conductivity of $0.05 \frac{W}{m \cdot K}$ and a unity initial velocity on the particle emission reduction η for various tube diameters.

3.1 Experimental verification

The analysis presented above has two major consequences for the design of the aerosol condenser: the trade-off between improving the deposition efficiency by reducing the overall heat transfer coefficient and the corresponding increase of the heat transferring surface has to be optimised, whereas the small dimensions of the heat exchanger passages require consideration of alternatives to the traditional shell and tube heat exchanger design. Furthermore it was deemed necessary to gain some practical experience with the obvious show stoppers for the application of a compact heat exchanger in a biomass boiler: fouling and removal of the condensate layers.

As an intermediate step towards a compact heat exchanger design, a 10 kW_{th} prototype heat exchanger was manufactured featuring removable plates and water cooling (figure 4). The plates are insulated using fibre board to reduce the heat transfer in a defined manner. The temperature profile inside the heat exchanger was calculated using an analytical model checked with a numerical approach (Best 2007). Experiments were conducted using a pilot scale biomass combustion plant with a nominal capacity of 180 kW_{th}. The biomass combustion plant itself consists of a furnace equipped with a horizontally moving grate, staged air injection and a flame tube boiler. The heat exchanger is again connected with the secondary combustion zone through a slip stream.

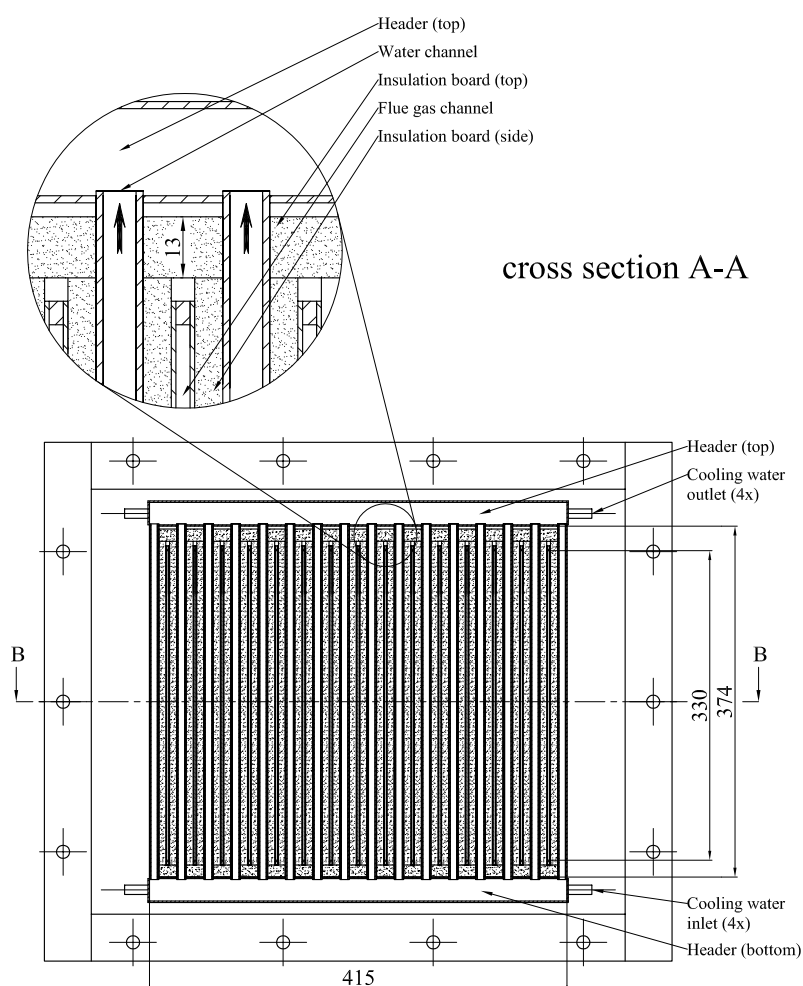


Figure 4: Cross sectional front view of the insulated plate type heat exchanger

Impactor measurements (figure 5) were performed at a flue gas temperature of 1000 °C to prevent major aerosol formation near the inlet and at 800 °C to simulate the worst case conditions that may occur in a small scale biomass combustion plant. The measurements at 1000 °C show a clear peak of the average PSD downstream the ordinary boiler at the 2nd impactor stage (cut-off diameter of 0.125 μm), this is consistent with earlier measurements reported in (Best 2007). The peak of the average PSD downstream the insulated plate heat exchanger is shifted towards a smaller cut-off diameter of the 0.0625 μm stage, see Fig. 5 and it can be concluded that a large fraction of the aerosol forming vapours is precipitated on the heat exchanger surfaces. No clear peak regarding the PSD can be distinguished for experiments

performed at secondary flue gas temperatures around 800 °C. The peak of the average PSD downstream the boiler is around the 3rd and 4th stage (cut-off diameter of respectively 0.25 μm and 0.5 μm). After the heat exchanger the peak can be observed around the second and third stage (0.125 μm respectively 0.25 μm). As expected the aerosol formation in the secondary combustion zone shows a significant increase compared to the measurements at 1000 °C, but the emissions after the heat exchanger are less than predicted by the model as deposition of already formed particles is also caused by thermophoresis (Best 2007).

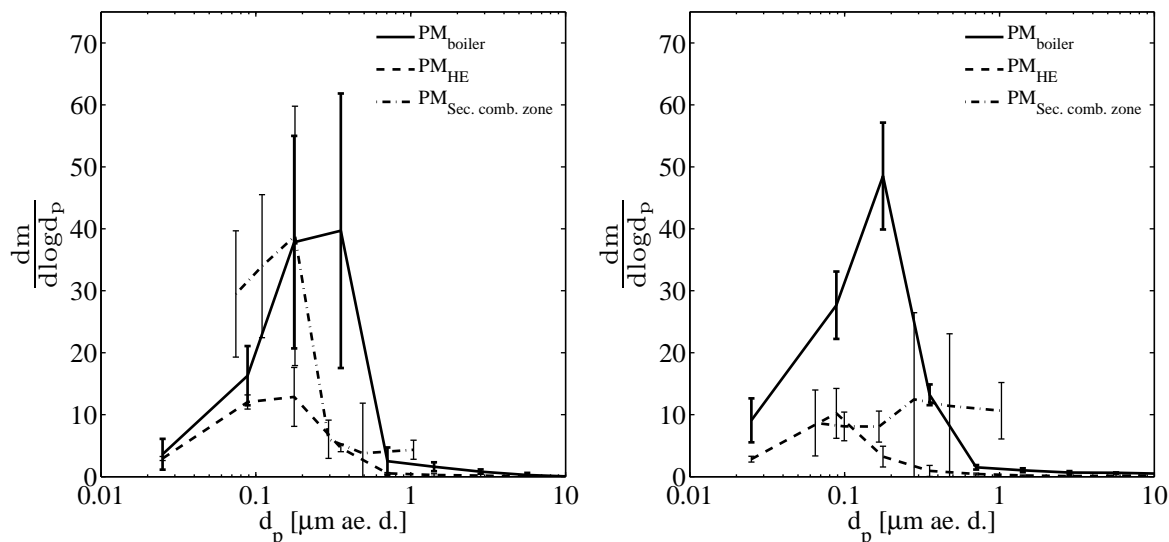


Figure 5: Average PSD downstream the ordinary boiler and the insulated heat exchanger (BLPI measurements) as well as inside the furnace (HT – LPI measurements) at secondary flue gas temperatures of about 800 °C (left) and 1000 °C (right).

4 Conclusions

With the insulated heat exchanger it was expected that a minimum fine particulate emission reduction of 65 % could be achieved at an initial flue gas temperature of about 1000 °C on the assumption that the potassium sulphate is the main aerosol forming compound. This assumption is backed by a fuel analysis showing that alkaline sulphates represents more than 80 wt. % of the total aerosol composition.

Using the insulated heat exchanger quite constant aerosol emission reductions could be achieved of about 80 ± 2 % on average when applying secondary flue gas temperatures of about 1000 °C. On average an aerosol emission reduction could be achieved of 64 ± 7 % when applying secondary flue gas temperatures of about 800 °C. The measured performance is better than predicted as the effect of thermophoresis is not included in the model.

With the results of the insulated plate heat exchanger, a commercially viable design has been made starting from a fin-tube layout as used in economisers. The temperature profile of the insulated plate heat exchanger is mimicked by introducing heat resistances. The prototype is currently undergoing testing at Graz University.

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