Influence of Dilution System and Electrical Low Pressure Impactor Performance on Particulate Emission Measurements from a Medium-scale Biomass Boiler

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Abstract

The accurate monitoring of particulate emissions from medium-scale, decentralized biomass combustion units is a major challenge for the deployment of this technology in the frame of the current energy transition. More specifically, the experimental characterization of the size of the emitted particles, i.e. using impactors, is still subject to discussions about the impact of the methodology on the measurement results. To meet with these challenges, particulate emissions from a medium-scale biomass boiler (4.5 MWth) were measured with Electrical Low Pressure Impactors (ELPI+) using two different dilution systems to examine the effect of dilution and performance of the ELPI+. For both the two stage dilution systems and one stage dilution system, no statistically significant correlation was found between the dilution ratio (DR) and particle total number concentration $N_{\text{tot}}$ or between DR and mass concentration $m_{\text{tot}}$. However, with both dilution systems, a significant positive correlation was observed between DR and particles with diameter $D_p < 0.01 \mu m$ due to measurement artefacts. With the one stage dilution system, condensation appears to be promoted with reduced DR. When the ELPI+ impactor is not overloaded, the number concentration of fractions $N_{<0.01}$ and $N_{0.01-0.1}$ reduce over time due to prematurely measuring particles. When the impactor is overloaded, $N_{<0.01}$ is overestimated and $N_{0.01-0.1}$ and $N_{0.1-1}$ are underestimated due to a change in cut-off diameter.

Keywords: Particulate matter; Biomass combustion; Dilution; ELPI+; Aerosol sampling

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INTRODUCTION

Biomass has been used for centuries around the world for generating energy due to its availability. More recently, biomass is gaining popularity due to being a viable alternative for fossil fuel but with a short-term carbon cycle, thus helping to reduce global warming (McKendry, 2002). Biomass combustion will not only avoid additional CO₂ emissions, but medium-scale biomass combustion for district heating is often economically viable (Soltero et al., 2018). Biomass will be among the most important energy sources of the future (Dhillon and von Wuehlisch, 2013).

A disadvantage of biomass combustion (in particular with solid fuels), beside NOx emission, is the emission of particles which have a known adverse effect on human health (Dockery et al., 1993; Kim et al., 2018). The health effects of inhalable particulate matter (PM) are respiratory and cardiovascular morbidity, such as aggravation of asthma, respiratory symptoms and an increase in hospital admissions; and mortality from cardiovascular and respiratory diseases and from lung cancer (WHO, 2013). Although PM is often expressed as mass concentration, research has shown that when exposed to the same mass concentration but with a different particle size, the smaller particles causes a greater adverse effect (Oberdörster et al., 1994) and thus, the particle size and particle concentration (or combined as the particle number size distribution) may be more appropriate to quantify PM. When investigating the toxicity of inhalable nanoparticles,
particle surface area can be expected to be the most relevant metric to quantify PM (Schmid and Stoeger, 2016).

PM resulting from biomass combustion can be divided into organic and inorganic pollutants. Organic PM consist of products from incomplete combustion (PIC) and can be reduced by improving the combustion efficiency, while inorganic PM is mainly derived from ash constituents in the biomass (Nussbaumer, 2003). Various factors such as combustion conditions, type of biomass and combustion appliance have an effect on the particulate emission (McDonald et al., 2000). In medium- and large-scale boilers, organic PM contributes only a few percentages to PM$_1$ although when the combustion conditions deteriorate, the organic fraction can become more dominant (Wierzbicka et al., 2005). Due to boiler design and combustion conditions the total polycyclic aromatic hydrocarbons (PAH) concentrations may vary over almost three orders of magnitude (Lillieblad et al., 2004). Depending on the combustion conditions and the type of combustion appliance, the particulate emissions from small-scale combustion appliances, expressed as mass concentration may vary up to a factor of 350 (Amaral et al., 2016) while the particle number concentration can vary in the range of $10^7$-$10^{10}$ particles per cm$^3$ (Johansson et al., 2003). For medium- and large-scale installations, the particulate emissions emitted into the atmosphere mainly depend on the used particle removal technology such as fabric filters and
electrostatic precipitators. However, the collection efficiency of these technologies are dependent on the particle size (Strand et al., 2002; Hasler and Nussbaumer, 1999).

An aerosol may contain up to three modes of particles: a nuclei mode (0.005-0.1 µm), an accumulation mode (0.1-2 µm) and a coarse mode (> 2 µm) (Whitby, 1978; John, 2011). With medium-scale biomass boilers, the fine mode (i.e. particles smaller than the coarse mode) is usually dominant over the coarse mode in both total number concentration and total mass concentration (Wierzbicka et al., 2005). During combustion of woody fuels in medium-scale boilers, the dominant elements in the coarse mode are Ca, K and S (Pagels et al., 2003; Wierzbicka et al., 2005) but also Mg, Si and Al are present (Jöller et al., 2005). The primary particles in the coarse mode mainly consist of compounds with low volatility such as CaO or SiO$_2$ but the mode also contains the not evaporated part of volatile compounds such as compounds of K and S (Jöller et al., 2005). The composition of the fine mode depends on the fuel composition due to the different formation pathway, as explained by Jöller et al. (2005): For wood chips, the fine mode consists of mainly K, S and Cl gaseous compounds which undergo nucleation (due to the lack of seed particles) and subsequent condensation on particle surfaces. For bark, a considerable amount of fine Ca-rich particles, which act as seed particles for the condensation of alkali sulphates and chlorides, are released during combustion along with K, S and Cl. These Ca-rich particles may act as seeds for condensation and therefore the nucleation
rates of new particles might be decreased. For waste wood, the main elements in the fine mode are Na, K, Cl, S, Zn and Pb. Zn is expected to evaporate to subsequently form ZnO, which nucleates and forms the seed particles for all further condensable species. Nucleation of other elements seems to be suppressed by surface condensation on these ZnO particles. Christensen and Livbjerg (1996) found that in the case of straw combustion, the fine mode mainly consists of KCl and K$_2$SO$_4$ (formed by homogeneous nucleation) and in minor amounts of P while the elements Ca, Mg and Si were detected in the coarse mode. If the amount of K in the flue gas exceed the sum of Cl and S, the excess K exists as gaseous KOH which might condense as either K$_2$CO$_3$ or KOH (Christensen and Livbjerg, 1996). This is usually not the case with straw combustion but this can occur during woody biomass combustion (Christensen and Livbjerg, 1996; Valmari et al., 1998).

Aerosols are unstable by nature and are therefore challenging to measure unequivocally. Changes in size or composition of particles involve heat and mass transfer between the particles and with the surrounding gas, for example by condensation, evaporation, nucleation, adsorption, absorption, coagulation and chemical reaction. Chemical reactions within a particle, between particles and with the surrounding gas may be non-growth processes that change the composition or density of the aerosol particle but with little or no change in particle size (Hinds, 2011).
To obtain a representative measurement of an aerosol, the sampling system that collects the sample must be designed to maximize the quenching of aerosol chemistry and dynamics (nucleation, coagulation and condensation) in order to minimize modification of the aerosol size distribution in the sampling system (Biswas and Thimsen, 2011). This can be performed by diluting the flue gas sample with particle- and oil-free dry instrument air before characterizing the particulate emissions. Dilution of the sample reduces the particle concentration and flue gas temperature in order to stop chemical reactions in the gas flow and prevents further variation of the particle size in the sampling system thus providing a more realistic and representative sampling of PM emissions (Amaral et al., 2015). Furthermore, most instruments that measure particle concentrations require dilution to obtain a sample that is within the measuring range of the measuring device. In addition, dilution prevents the condensation of water when the flue gases are cooled. However, research on internal combustion engines has shown that the physical and chemical properties of exhaust aerosols continuously change after their formation and one may significantly modify the aerosol properties by controlling this dilution process and thereby affecting the measurement (Giechaskiel et al., 2014). The particle size and concentration of volatile material emitted by diesel engines are strongly dependent upon dilution and sampling conditions, because sulphur and organic compounds are generally in the vapour phase in the tailpipe, and undergo a gas-to-particle conversion during dilution and cooling (Kittelson et al.,
The optimum dilution system for an aerosol depends strongly on the focus of the measurement (Lyyränen et al., 2004). The aim may be to simulate the dilution effect when the flue gas enters the atmosphere (i.e. atmospheric dilution) or the aim may be to preserve the number size distributions as unbiased as possible from the measurement and dilution setup artefacts and to produce a so called ‘real’ size distribution of the particles as observed inside the chimney (Lyyränen et al., 2004). With atmospheric dilution, also the formation of secondary organic aerosols (SOA) due to photochemical oxidation of volatile organic compounds (VOC) need to be accounted for since these emissions can cause the total organic aerosol concentration to increase by a factor of 1.5 to 2.8 (Grieshop et al., 2009).

In this study, the particle number size distribution of a medium-scale biomass boiler was measured using two Electrical Low Pressure Impactors (ELPI+) simultaneously with each a different compact partial flow dilution system: a porous tube diluter in combination with an ejector diluter (PTD+ED) and a Dekati axial diluter (DAD 100). The first objective is to evaluate the effect of the dilution system on the particles number size distribution. Both dilution systems have a heated dilution stage where the vapour pressure is reduced and thus the gas-to-particle transitions should be minimized which should lead to a representative particle number size distribution of the particulate emissions in the chimney. The second objective is to study the performance of the ELPI+ under different particle loads and under different boiler configurations.
METHODS

Plant description

Parallel measurements were carried out at a 4.5 MWth biomass grate boiler that provides district heating to a nearby residential area. Downstream of the boiler, the plant is equipped with a multi-cyclone, a baghouse filter and a 0.83 MWth flue gas condenser (FGC), with the possibility to bypass the baghouse filters and the FGC separately. A schematic overview of the plant is given in the supplementary material Fig. S1. The influence of the FGC on the emissions was not the focus of this study and was therefore bypassed during the entire measuring period. The sampling probes for extracting the flue gas samples were located between the baghouse filters and the FGC. The flue gas temperature was around 150°C. The boiler was fueled with wood chips with a water content of about 45-50% (based on wet mass) originating from forests within an area of 100 km.

Sampling systems

The sampling setup is shown in Fig. 1. Two Dekati electrical low pressure impactors (ELPI+) systems were used in parallel, both with a different compact partial flow dilution system, to measure the particle number size distribution of particles with a diameter between 6 nm and 10 µm, divided in 14 size fractions. The operating principle of the ELPI+ is based on the electric charging of the particles using a corona charger, after which the particles are classified in a cascade of inertial impactors that can detect the particle’s electrical charge. A more thorough
A description of the ELPI+ and its performance is given by Järvinen et al. (2014). A third sampling port was used to analyze the gaseous compounds in the flue gas. Air was used as a dilution gas to reduce particle and gas concentrations and to lower the temperature of the flue gas sample before entering the ELPI+. The dilution air originated from compressed air from the plant that underwent an air treatment to make it particle- and oil-free dry air. Each dilution system had its own air treatment unit.

In both dilution systems the loss of particles is estimated to be low. Diffusional deposition is minimized in both dilution systems because the dilution air is introduced as a layer around the flue gas samples. Both gravitational settling and inertial depositions are considered of being of minor importance since no coarse fraction is present in the flue gas after the multi-cyclone and these loses reduce with reducing particle diameter (Brockmann, 2011). Thermophoretic deposition is low since both dilution systems use heated air meaning that the deposition is reduced due to the lowered temperature gradient. Although, there is a difference between the temperature used in the two dilution systems.

In order to improve the particle collection efficiencies of the ELPI+ impactors, grease was applied on the aluminum collection substrates (Dzubay et al., 1976; Kuuluvaivanen et al., 2017; Dunbar et al., 2005). Not greasing the substrates results in a larger bouncing effect of impacted particles from a higher impactor stage to a lower stage which leads to an underestimation of the
number of particles in the correct size fraction and an overestimation of the number of particles in a smaller size fraction (Virtanen et al., 2010; Chen et al., 2011). Greased substrates initially have a good collection efficiency but this decreases with particle load in the impactor (Turner and Hering, 1987; Tsai and Cheng, 1995). On the other hand, with ungreased substrates, the particle load on the substrates changes the surface characteristics causing the collection efficiency to increase over time (Tsai and Cheng, 1995). With ELPI+ A the grease was dissolved in acetone and then applied with a brush on the substrates while with ELPI+ B the grease was applied as an aerosol using a spray can. When grease is applied with a brush, it is a thicker layer than when it is applied with a spray can. A thicker grease layer greatly improves the particle collection efficiency (Pak et al., 1992). A sample with a higher relative humidity will also increase the collection efficiency (Chen et al., 2011).

The porous tube diluter + ejector diluter system (PTD + ED) is a two stage dilution system manufactured by Venacontra (Fig. S2) and was connected to ELPI+ A. The purpose of the two staged system is to keep the particle size distribution unchanged by first diluting the flue gas sample at the same temperature to lower the vapour pressure, and then decreasing the temperature by diluting at ambient temperature. In the first stage, the flue gas sample is introduced by a heated probe (at 150°C) in the PTD where it is mixed with heated (150°C) dilution air to lower the partial pressure while preserving the temperature. The heated air creates an unfavorable
environment for nucleation (Lyyranen et al., 2004) and condensation of semi-volatile organic compounds will be prevented (Fachinger et al., 2017). The PTD is a coaxial cylindrical diluter where the dilution air flows through a porous inner tube, thus creating a layer of clean air around the aerosol sample (Lyyranen et al., 2004; Auvinen et al., 2000). The flow velocity of the dilution air through the pores is greater than the deposition velocity of the particles, thus the particles will not deposit in the PTD (Auvinen et al., 2000). In addition, the use of heated dilution air will prevent particles from depositing due to thermophoresis. In the second stage, the flue gas sample is mixed in the ED with dilution air at ambient temperature in order to further dilute the sample and to reduce the temperature before entering the ELPI+. The ED ensures good mixing and rapid entrainment because the flue gas sample enters the mixing chamber through a venturi with a high velocity (Lyyranen et al., 2004). The PTD + ED dilution system is widely used to sample particulate emissions from biomass combustion (Tissari et al., 2007, 2008b; Dyakov et al., 2016; Kaivosoja et al., 2013; Grigonyte et al., 2014; Lamberg et al., 2011; Torvela et al., 2014). The calculation of the dilution ratio (DR) is based on the dry CO₂ concentration before and after dilution corrected for the background CO₂ (Grigonyte et al., 2014).

The Dekati axial diluter system (DAD 100) is a single stage dilution system (Fig. S3) and was connected to ELPI+ B. In the DAD 100, heated (60°C) dilution air is introduced as a layer around the flue gas sample resulting in low particle losses, as claimed by the manufacturer (Dekati,
The DAD 100 was used in previous research to dilute flue gases from coal combustion (Mertens et al., 2014, 2015). The DR is calculated with the volumetric flow rate of the dilution air and the diluted flue gas sample. Both calculation methods of DR (CO₂ based and flow based) result in the same definition of DR, i.e. ratio of the final flow to the initial flow. No dilution corresponds with a dilution ratio of one.

Three different gas analyzers were used to analyze the gaseous compounds of the flue gas. A Fourier-transform infrared spectroscopy (FTIR) analyzer was used to monitor the concentration of water vapour, carbon dioxide, carbon monoxide, nitrogen monoxide, nitrogen dioxide, nitrous oxide, sulphur dioxide, hydrogen chloride, hydrogen fluoride, ammonia and organic gaseous compounds (OGC). The FTIR was equipped with a zirconium dioxide (ZrO₂) sensor to measure the oxygen concentration. A flame ionization detector (FID) was used to measure the total organic carbon (TOC) and was calibrated with propane. The flue gas was sampled at 180°C and was kept constant throughout the FTIR, the FID and the sampling lines in between, to allow the analysis of the hot and wet flue gas (and of the less volatile organic compounds in it) and to prevent condensation thus avoiding loss of water soluble components. A second outlet of the FTIR was connected with a gas cooler in order to dry and cool the sample after which the third gas analyzer, a Horiba PG-250, measured the dry oxygen, carbon dioxide, carbon monoxide,
nitrogen oxide and sulphur dioxide concentrations. The Horiba was calibrated with the appropriate zero and span gases.

Measurement plan

The emissions of combustion were studied with multiple tests at three different operation modes of the plant: boiler at high load (i.e. 100%), boiler at low load (i.e. 50%) and boiler at high load with bypass of the baghouse filters, denoted by respectively HL, LL and HL BB. The measurements were carried out with both ELPI+’s and then the measured concentrations were compared with each other. ELPI+ A is therefore the reference for ELPI+ B and vice versa. Using another measuring device as a reference, such as a scanning mobility particle sizer (SMPS), is not as straightforward because the aerodynamic diameter and mobility diameter can be extremely different (van Gulijk et al., 2004). The tests were carried out on two consecutive days, namely S1 and S2. During the first day, the boiler was operated continuously at high load, which allowed to compare the ELPI+’s, compare the dilution setups and study the influence of DR. The second day the boiler was operated first at high load, then with bypass of the baghouse, then at low load and finally back to high load. These tests allowed to investigate the importance of greasing the ELPI+ substrates, the influence of the boiler operation mode and the influence of overloading the ELPI+ impactor. Due to the high number of particles in the flue gas during the bypass of the baghouse, both ELPI+ impactors were overloaded. The user manual of the ELPI+ states that a total of 1 mg
of particles collected per impactor stage can be considered as an absolute maximum although this value can depend on the particle distribution and the particle properties (Dekati, 2016). After the baghouse bypass tests, a clean impactor was inserted in ELPI+ B while with ELPI+ A the overloaded impactor was retained for the remaining tests. This allowed for comparing the influence of overloading the ELPI+. Garra et al. (2016) concluded that long term collection with the original ELPI biased the particle size distribution by measuring more particles in stages 6-8 and less particles in stages 3-5. The authors explained the biasing by the change in jet-to-impaction distance due to the formation of domes of accumulated particles on the impactor which changes the cut-off diameter of the impactor. Because the ELPI+ contains an additional impactor stage and an electrical filter stage (Järvinen et al., 2014), this would correspond with more particles expected in stages 8-10 and less in stages 5-7 as a bias from long term collection with the ELPI+. Impactors have been designed and developed that can reduce and even eliminate the effects of particle overloading (Tsai and Cheng, 1995; Peters et al., 2001; Tsai et al., 2012; Chien et al., 2015; Le and Tsai, 2017; Le et al., 2019), but these are not suitable for implementation in the ELPI+. It should however be noted that the effects of overloading the ELPI+ impactor can be largely postponed by using oil-soaked sintered impactor plates instead of greased impactor plates (van Gulijk et al., 2003).
The ELPI+ measures in real-time and therefore the ELPI+ results are time averaged values (± standard deviations) of each test. The average sampling duration for each ELPI+ test was 10 minutes with a minimum of 6 minutes and a maximum of 18 minutes. Tests shorter than 6 minutes are excluded from the analysis. As is the case with the ELPI+, the gas analyzers measure in real-time and therefore the results are also time averaged values (± standard deviations) of each test.

The average particle density $\rho$ is an input parameter of the ELPI+ and has to be estimated, although the chemical compositions of different size fractions may induce a significant change in density per particle size (Coudray et al., 2009). Nonetheless, since the particulate emissions are dominated by the fine particles, the average particle density from wood combustion was estimated at 2.0 g cm$^{-3}$ as assumed by Pagels et al. (2002) and Lillieblad et al. (2004) and as concluded by Coudray et al. (2009). With both dilution systems, isokineti sampling was not necessary because the flue gas is dominated by fine particles, as almost all coarse particles are trapped by the multi-cyclone and thus the particles follow the gas streamlines (Hinds, 1999).

RESULTS AND DISCUSSION

The PM emissions are characterized by their particle number size distribution which can be summarized by the quantities total number of particles $N_{\text{tot}}$ and geometric mean particle.
diameter (GMD). The dilution settings of the different tests and \( N_{\text{tot}} \) and GMD are represented in Table 1. The displayed ELPI+ results are corrected for their dilution. Some data were rejected from the analysis due to incorrect or incomplete measuring with the ELPI+ (see notes in Table 1).

The two dilution systems are complementary since the DAD 100 operates with a DR between 4 and 10 while the PTD + ED operates with a DR between 10 and 51. With the PTD + ED the DR is calculated as a time averaged value (± standard deviation) from continuous CO\(_2\) measurements. With the DAD 100 the DR was set to a specific value and was not measured continuously and so there is no standard deviation. The individual DR for PTD and ED are estimated using the known volumetric flow rates, the value of DR\(_{\text{tot}}\) and the formula DR\(_{\text{tot}}\) = DR\(_{\text{PTD}}\) × DR\(_{\text{ED}}\). The residence time of the particles in the PTD and the ED were estimated in the ranges 0.3-0.5 s and 0.8-0.9 s, respectively. The residence time in the DAD 100 dilution system can not be estimated unequivocally as dilution takes place in the extension of the sampling line and thus the mixing chamber is not clearly defined.

The results of the gas analyzers for each test are shown in Table 2. All gas concentrations are dry based (except for H\(_2\)O) and are corrected to a 13 vol% O\(_2\) concentration (except for H\(_2\)O and O\(_2\)) using the H\(_2\)O and O\(_2\) concentrations measured with the FTIR. The Horiba gas analyzer was able to measure dry concentrations of O\(_2\), CO\(_2\), CO, NO\(_x\) and SO\(_2\) but the SO\(_2\) concentration was below the detection limit (ppm level). The TOC values measured by the FID are expressed as C-
equivalent mass per normal cubic meter dry gas, corrected to a O$_2$ concentration of 13 vol%. The measured TOC concentrations are low (<5 mg C Nm$^{-3}$) and close to the estimated limit of quantification of the FID. Besides the compounds presented in Table 2, the FTIR was also able to monitor N$_2$O, SO$_2$, HCl, HF, NH$_3$, CH$_4$, acetaldehyde and formaldehyde, and public available spectra were used to detect formic acid and acetic acid but none of these components were detected above the detection limit (ppm level). The O$_2$ measured with the FTIR appears to be slightly higher than the O$_2$ concentration measured with the Horiba. This difference may be due to the fact that the FTIR results are obtained by measuring in the wet flue gas and then calculating the concentration on dry basis using the measured H$_2$O concentration while the Horiba measures the concentration in the dried flue gas. Additionally, the difference may be due the difference in O$_2$ measuring method as the FTIR uses a zirconium cell while the Horiba uses a galvanic cell. These small difference make that some of the other gas concentrations are slightly different. The CO$_2$ concentration appears to be slightly higher with the Horiba. The CO concentration appears to be higher with the Horiba the first tests and higher with the FTIR the last tests but both show an increased concentration during low boiler operation and they have similar standard deviations. The NO$_x$ concentration is lower with the FTIR but both analyzers show a decreased concentration during low boiler operation which may be related to the lower temperature in the combustion chamber.
Time series of particle concentrations

One of the main advantages of the ELPI+, in addition to its ability to measure a large range of particle sizes, is that it has a fast sampling rate (10 Hz). This allows for a continuously measurement the particle number concentration in each impactor stage. In Fig. 2 the time series of particulate and gaseous emissions during the first measuring day are presented. In Fig. 2a the flue gas was diluted with the PTD + ED and in Fig. 2b the DAD 100 dilution system is used. The evolution in time of the total number of particles \( N_{\text{tot}} \) and size fractions \( N_{0.1-1} (0.1 \, \mu\text{m} < D_p < 1 \, \mu\text{m}: \text{stages 5-9}) \), \( N_{0.01-0.1} (0.01 \, \mu\text{m} < D_p < 0.1 \, \mu\text{m}: \text{stages 2-4}) \) and \( N_{<0.01} (D_p < 0.01 \, \mu\text{m}: \text{stage 1}) \) are presented. The size fraction \( N_{1-10} (1 \, \mu\text{m} < D_p < 10 \, \mu\text{m}: \text{stages 10-14}) \) is not presented here since the amount of particles measured in this fraction in several order of magnitude lower than in the smaller fractions. In Fig. 2c the gas concentrations in the flue gas measured by the FTIR are presented. The gas concentrates are dry based and corrected to 13 vol% \( \text{O}_2 \). The gas concentrations indicate that the boiler operation during high load is quite stable over a large time scale. Over a smaller time scale, variations can be observed in most gas concentrations. These variations appear to correspond to the variations in particulate emissions.

The PTD + ED dilution system was connected to ELPI+ A and the DAD 100 dilution system was connected to ELPI+ B during all tests, except during test S1.2. During test S1.2, the ELPI+’s were interchanged to check whether they yield the same results with the same dilution system (i.e.
the DAD 100 dilution system was connected to ELPI+ A and the PTD + ED dilution system was connected to ELPI+ B). All measurements performed with ELPI+ A are displayed as solid lines while all measurements performed with ELPI+ B are displayed as dashed lines. Seen from the PTD + ED dilution system point of view (Fig. 2a), there is a good agreement between the two ELPI+’s, although $N_{\text{tot}}$ is slightly higher compared with the previous and next test. This is mainly the result of the higher amount of particle in fraction $N_{0.01-0.1}$. Seen from the DAD 100 dilution system point of view (Fig. 2b), there is a relative good agreement between the two ELPI+’s. Fraction $N_{0.1-1}$ during S1.2 is similar to the other tests. Fraction $N_{0.01-0.1}$ during S1.2 is situated between the previous and next test with which it follows the trend of the DAD 100 that this fraction decreases with time. Fraction $N_{<0.01}$ with the DAD 100 during S1.2 appears to have two peaks which corresponds well with the peaks in $N_{\text{tot}}$ of the PTD + ED although with the PTD + ED these peaks are visible in both $N_{<0.01}$ and $N_{0.01-0.1}$. Based on these observations, it can be concluded that both ELPI+’s yield the same results, although there is a relatively large variation between the measurements with the same ELPI+ over time. These variations over time are believed to be the result of the increasing effect of prematurely measuring the smallest particles, as argued by van Gulijk et al. (2001). The authors propose that this phenomenon occurs in three phases (van Gulijk et al., 2001): First, at very low particle load on the impactors, the impactor stages of the accumulation peak (ELPI+ stage 5 and 6) are covered with a fluffy bed of
particles, which acts as a filter for the smaller particles. Then, in a second phase, at slightly higher impactor load, piles of particles form on the impactor plates and thereby change the cut-off diameter. In a third phase, at high impactor load, the piles of particles have become so large that re-entrainment occurs and probably a mechanical equilibrium develops. Fraction $N_{0.01-0.1}$ appears to reduce over time with both dilution systems although the gas concentrations do not suggest that the combustion conditions change. This suggests that the ELPI+ underestimates the amount of particles in stages 2-4 with increasing time, even when the ELPI+ impactor is not overloaded. This can be explained by the phenomenon of prematurely measuring those particles. With the PTD + ED, fraction $N_{0.01-0.1}$ is slightly above fraction $N_{0.1-1}$ during the first two tests and is slightly below fraction $N_{0.1-1}$ during the last two tests. With the DAD 100 this trend is more pronounced as the used DR is lower and thus the particle load on the impactor of ELPI+ B is larger. During test S1.1 with DAD 100, fraction $N_{<0.01}$ is 1.8 times higher and fraction $N_{0.01-0.1}$ is 4.8 times higher than during test S1.3. Both fractions further reduce over time. Fraction $N_{<0.01}$ also appears to have a positive correlation with DR, as will be discussed later on. Particle fraction $N_{0.1-1}$ appears to be similar and stable for both dilution systems and does not seem to be affected by DR.

In Fig. 3 the time series of particulate and gaseous emissions during test S2.1 to S2.3 and S2.10 are presented. Mind the time gap between tests S2.3 and S2.10 because the tests at different
operation modes are discussed separately. During tests S2.1 with the PTD + ED (Fig. 3a) no grease was applied on the aluminum collector substrates of the ELPI+ impactor. \( N_{\text{tot}} \) and \( N_{<0.01} \) during this ‘no grease’ test are drawn relative to the grey scale since they are of a different order of magnitude due to the bouncing effect. In all other tests (including tests at other operation modes), grease was applied on the substrates. The purpose of this test is merely to demonstrate the importance of proper greasing and is therefore not included in the further analysis. With the ‘no grease’ test, the average \( N_{\text{tot}} \) was a factor of 4 higher and the average number of smallest particles, measured in the fraction \( N_{<0.01} \), was a factor of 10 higher compared with tests with the same dilution system when grease was applied. Fraction \( N_{0.01-0.1} \) during the ‘no grease’ test is mostly lower than test S2.3 with the PTD + ED and test S2.1 with the DAD 100 although the fraction reduce over time with all tests. The underestimation of the number of particles in the correct size fraction, due to the bouncing effect, is more difficult to observe because this is less pronounced. One particle not measured in the correct stage due to bounce is measured as multiple particles in a lower stage since the particle charging is size dependent. What makes the underestimation even more challenging to observe, is the variation of the size fractions over time. \( N_{\text{tot}} \) decreases from \( 1.3 \times 10^7 \) particles per cm\(^3\) at the start of the test to \( 2.9 \times 10^6 \) particles per cm\(^3\) at the end of the test, which is mainly the result of the decreasing of fraction \( N_{<0.01} \). Also fraction \( N_{0.01-0.1} \) appears to decrease over time, especially at the start of the test. On the other hand,
fraction $N_{0.1-1}$ appears to increase over time at the start of the test. This is in accordance with what van Gulijk et al. (2001) observed: a decrease of particles with $D_p$ between 0.03 and 0.1 µm and an increase of particles with $D_p > 0.1$ µm over time. This phenomenon of prematurely measuring small particles causes the opposite effect than the particle bounce: instead of particles travelling too far there are particles that travel not far enough in the impactor (van Gulijk et al., 2001). The effect of prematurely measuring small particles can also be observed during test S2.1 to S2.3 with the DAD 100 dilution system (Fig. 3b) as both fractions $N_{<0.01}$ and $N_{0.01-0.1}$ decrease over time. This effect starts almost immediately at the start of measurement S2.1 (i.e. with a clean impactor) and increases over time. The underestimation of fractions $N_{<0.01}$ and $N_{0.01-0.1}$ and overestimation of fraction $N_{0.1-1}$ due to the prematurely measuring of particles increase over time but become more stable after a while due to an equilibrium that develops in the impactor. With the DAD 100, an increase of the fraction $N_{0.1-1}$ is however not observable. It is noted that during the measurements of tests S2.2 and S2.3 with the DAD 100 dilution system an unstable pressure was observed in the ELPI+ which can be the reason for the fluctuations in fraction $N_{<0.01}$. For tests S2.1 to S2.3 and S2.10 with the DAD 100 and test S2.3 with the PTD + ED fraction $N_{0.1-1}$ appears to be similar for both dilution systems and similar to S1. Fraction $N_{<0.01}$ with the PTD + ED dilution system during test S2.3 is similar to the one of test S1.4 which has almost the same DR, although during test S2.3 an increasing trend can be observed. During test S2.2 with the
PTD + ED fraction $N_{0.01}$ is higher as expected. Since this test is the first test with a clean impactor (with greased substrates), the effect of prematurely measuring particles is not yet so pronounced. However, a decreasing trend can not be observed due to the limited duration of the test. Test S2.10 with the PTD + ED was performed with an overloaded ELPI+ impactor. Both $N_{0.01-0.1}$ and $N_{0.1-1}$ appear to be underestimated compared with the DAD 100 and $N_{<0.01}$ appears to be overestimated. The underestimation may be due to the change in cut-off diameter of the impactor stages and the overestimation may be the result of an increasing bouncing effect due to the saturated impactor plates. Note that, based on the gas concentrations, the boiler is still recovering from the low load operation mode and is not yet fully stable (Fig. 3c).

In Fig. 4 the time series of particulate and gaseous emissions during test S2.4 to S2.5b are presented. During these tests, the boiler was operated at high load with the baghouse filters bypassed. During this operation mode, $N_{\text{tot}}$ is much higher and fraction $N_{0.1-1}$ is clearly more dominant, demonstrating the need for the installation of baghouse filters. The accumulative mass of particles collected per impactor stage during these tests exceeded the limit value of 1 mg given by the ELPI+ user manual (Dekati, 2016). With the PTD + ED during test S2.4 stages 6-8 and 13-14 got overloaded and by the end of test S2.5b stages 6-14 were overloaded. With the DAD 100 stages 6-9 and 13-14 were overloaded by the end of test S2.5a. With both dilution systems, fractions $N_{0.1-1}$ and $N_{0.01-0.1}$ reduce over the time. This is in accordance with overload test S2.10.
with the PTD + ED, as the overload increases, the concentrations of fractions $N_{0.1-1}$ and $N_{0.01-0.1}$ are increasingly underestimated. With the DAD 100 fraction $N_{<0.01}$ also reduces over time. However, with the PTD + ED fraction $N_{<0.01}$ does not appear to reduce over time but appears to correlate with DR.

In Fig. 5 the time series of particulate and gaseous emissions during test S2.6 to S2.9 are presented. During these tests, the boiler was operated at low load, which can be observed from the increased CO concentration and the reduced NO$_x$ concentration (Fig. 5c). Note that the combustion conditions are not fully stable as the CO concentration is still increasing and the NO$_x$ concentration is still decreasing during the first two tests. The impactor of ELPI+ A, connected to the PTD + ED dilution system, is still overloaded with particles due to the previous tests. A clean impactor with greased substrates was inserted in ELPI+ B. With the PTD + ED dilution system, $N_{0.1-1}$ and $N_{0.01-0.1}$ appear to reduce over time (Fig. 5a). Fractions $N_{0.1-1}$ and $N_{0.01-0.1}$ are believed to be more underestimated as the overloading increases because of a shift in cut-off diameter. Moreover, fraction $N_{<0.01}$ appears to correlate with DR. With the DAD 100 dilution system, fraction $N_{0.1-1}$ appears to be stable and fractions $N_{0.01-0.1}$ and $N_{<0.01}$ appear to reduce over time during test S2.8 and S2.9 (Fig. 5b).

**Particle number size distributions**
The PM emissions measured by the ELPI+ are expressed as time series of particle number concentrations for each impactor stage. When time-averaging the particle concentration in each stage, an average particle number size distribution for each test can be obtained. This distribution represents the amount of particles measured within a size interval divided by the logarithmic range of the size interval (dN/d log \(D_p\) on vertical axis) at the geometric mean of each size interval (\(D_p\) on horizontal axis).

Before going deeper into the different operation modes, observations are made about the general shape of the distributions (Fig. 6-9). With both dilution systems and during all operation modes, an accumulation mode with a peak between 0.1 and 0.5 \(\mu\)m can be distinguished, which corresponds with previous research (Krugly et al., 2014; Lamberg et al., 2011; Sippula et al., 2007; Johansson et al., 2003; Tissari et al., 2008b; Hueglin et al., 1997; Fachinger et al., 2017; Pagels et al., 2003). With most of the tests, the smallest particles (\(D_p < 0.03 \mu\)m: stages 1 and 2) suggest that also a second mode may be present, although this has to be interpreted with the necessary caution as the measuring range of the ELPI+ does not completely cover this mode. The observation of a nuclei mode in the particle number size distribution has been reported in literature regarding residential wood combustion (Krugly et al., 2014; Fachinger et al., 2017; Hedberg et al., 2002). However, with medium-scale boilers, the observation of a nuclei mode is rather unusual although it has been observed in some work as a weak mode around 40 nm (Pagels
et al., 2003). Therefore, the relative high number of the smallest particles ($D_p < 0.03 \mu m$) is probably the result of measuring artefacts and the result of the baghouse filters that have a lower collection efficiency for those particles, which both will be discussed later on.

In Fig. 6 and 7 the particle number size distributions at high load with each of the two dilution systems are shown at different dilution ratio’s. With the PTD + ED dilution system, eight tests are included while with the DAD 100 dilution system nine tests are included. During test S1.2 (green distribution in both graphs), the ELPI+’s were interchanged (i.e.) to check whether they yield the same results with the same dilution system. Seen from the PTD + ED dilution system point of view (Fig. 6), there is a good agreement between the two ELPI+’s, except for impactor stage 2 ($D_p = 0.021 \mu m$) which shows a slightly higher amount of particles at test S1.2. Seen from the DAD 100 dilution system point of view (Fig. 7), there is also a good agreement between the two ELPI+’s, but a wide spread between the measurements with the original ELPI+ B can be observed (especially for particle with a diameter $D_p < 0.1 \mu m$). These variations between the measurements of ELPI+ B are believed to be the result of the increasing effect of prematurely measuring the smallest particles. It can be concluded that both ELPI+’s yield the same results, given the relatively large variation between measurements with the same ELPI+ over time, which is in accordance with what was already determined when observing the time series.
With the PTD + ED dilution system (Fig. 6), it can be observed that during the ‘no grease’ (n.g.) test (i.e. S2.1) the particles measured in the ELPI+ filter stage (i.e. stage 1) is clearly higher and the particles measured in stages 3 and 4 is slightly lower which is the due to the increased bounce effect: overestimation of small particles and underestimation of correct size. During the overload (o.l.) test (i.e. S2.10), the distribution clearly differs from the other tests. More particle can be observed in stage 1 and less in stages 2 to 6. The amount of particles in fraction $N_{0.01-0.1}$ is close to zero and the peak of the accumulation mode is halved. For the remaining high load tests with the PTD + ED dilution system, $N_{\text{tot}}$ is between $1.1 \times 10^6$ and $1.6 \times 10^6$ particles per cm$^3$ and GMD is between 27 nm and 40 nm. With four tests the peak of the accumulation mode is at $D_p = 0.12 \mu m$ (stage 5) and with two tests the peak is at $D_p = 0.20 \mu m$ (stage 6). The residence time during high load was estimated between 0.41 s and 0.48 s in the PTD and between 0.86 s and 0.89 s in the ED. It can be observed that an increase in DR leads to an increase of the smallest particles, measured in the ELPI+ stage 1, although there is a wide spread between tests with the same DR.

With the DAD 100 dilution system (Fig. 7), an unstable pressure (u.p.) in the ELPI+ was observed during two tests (i.e. S2.2 and S2.3). During these tests, the amount of particles in stage 1 appears to be lower compared to the other tests. For the remaining high load tests with the DAD 100, $N_{\text{tot}}$ is between $3.9 \times 10^5$ and $1.4 \times 10^6$ particles per cm$^3$ and GMD is between 33 nm and
133 nm. Notice the large spread between these quantities compared with the PTD + ED dilution system. The peak of the accumulation mode is at \( D_p = 0.12 \mu m \) (stage 5) when DR = 10 but the peak shifts to the right (\( D_p = 0.20 \mu m \): stage 6) when the DR decreases. As with the PTD + ED dilution system, it can be observed that an increase in DR leads to an increase in the number of smallest particles, measured with the ELPI+ stage 1, but there is an even wider spread between tests with the same DR. This wide spread between the amount of smallest particles in stage 1 is the cause for the large spread between \( N_{tot} \) and GMD.

The particle number size distributions during low load are presented in Fig. 8. With the PTD + ED four tests are included and with the DAD 100 two tests are included. With the DAD 100 the ELPI+ impactor was not overloaded and it can be observed that the peak of the accumulation mode shifts from stage 5 during high load (when DR = 10) to stage 6 during low load. This is in accordance with what has been reported in literature: a shift of the peak towards the larger particles sizes with decreasing load (Johansson et al., 2003; Tissari et al., 2008a; Lillieblad et al., 2004; Torvela et al., 2014). With the PTD + ED the tests are performed with an overloaded ELPI+ impactor. More particles can be observed in stage 1 and the amount of particles in fraction \( N_{0.01-0.1} \) is close to zero, which is in accordance with the high load overload test (i.e. S2.10 in Fig. 6). However, during the low load overload tests, the peak of the accumulation mode is not halved (compared to the DAD 100 tests) but the peak is rather shifted
from stage 5 and 6 during high load to stage 7 during low load. This shift can be both the result of
the decreasing boiler load, as with the DAD 100, and the result of the impactor overloading
(i.e. change in cut-off diameter of impactor stages). Note that with the PTD + ED the number of
particles in stages 1, 4, 5 and 6 decrease with decreasing DR, although this could also be a
consequent of the increasing overloading as the lowering of DR is performed chronologically.

The particle number size distributions at high load with bypass of the baghouse of four tests
with the PTD + ED and two tests with the DAD 100 are shown in Fig. 9. The accumulation mode
is more dominant during this operation mode, showing the need for installing baghouse filters.
Due to the high number of particles, the impactors of both ELPI+’s were overloaded with
particles, according to the ELPI+ user manual (Dekati, 2016). With both dilution systems, the
peak of the accumulation mode decreases with increasing test number and thus the peak
decreases as time goes on. With the PTD + ED the peak of the mode also shifts from stage 6 to
stage 7 over time and the number of particles in stage 7 increases over time which shows that the
cut-off diameters change as the particle loading increases. With the DAD 100, stages 2 and 3 are
close to zero. As with all previous tests, an increase in DR seems to lead to an increase of the
smallest particles. During this operation mode, the particle number concentration measured in
stage 6 and 7 with both ELPI+’s are close to upper limit of the ELPI+ measuring range of these
stages. An analogue discussion about the particle mass size distribution can be found in the supplementary material.

Comparison of dilution systems

The visual comparison of (a) total particle number, (b) total particle mass and (c) geometric mean diameter between the PTD + ED and the DAD 100 dilution system is shown in Fig. 10. The solid line indicates the 1:1 relation and the dashed lines mark the ± 30% interval. The points represent time averages of a test measured by both dilutions systems simultaneously. These tests are: four tests at high load, the high load test with the ELPI+’s interchanged, the high load test with no grease used with ELPI+ A (connected to PTD + ED), the high load test with the impactor of ELPI+ A overloaded (connected to PTD + ED), two low load tests (with ELPI+ A overloaded) and two tests at high load with the baghouse bypassed (with both ELPI+’s overloaded).

When diluting with the PTD + ED dilution system, a higher \( N_{\text{tot}} \) is obtained during all tests, except with one high load test (Fig. 10a). When diluting with the PTD + ED dilution system, a higher \( m_{\text{tot}} \) is obtained during all tests, except with the ELPI+ interchange test and the high load overload test (Fig. 10b). Thus, with exclusion of the overload test, there is always more particulate mass collected with ELPI+ A. This could be the result of the thicker layer of grease applied on the substrates of ELPI+ A that improves the particle collection efficiency. The GMD with the PTD + ED dilution system is lower than with the DAD 100 dilution system for all tests.
except one high load test (Fig. 10c). This exception is the same test as the exception in $N_{\text{tot}}$, namely test S1.1.

**Influence of dilution conditions on particulate measurements**

The influence of dilution on the number of particles, the mass of particles and the particle diameter is assessed using the Spearman correlation coefficient $r_s$. The correlations between DR and (i) number of particles $N$ per size fraction, (ii) mass of particles $m$ per size fraction, (iii) each stage separately and (iv) GMD are shown in Table 3. Four types of configurations had a sufficient amount of tests to perform the analysis: PTD + ED high load ($n = 6$), DAD 100 high load ($n = 7$), PTD + ED low load ($n = 4$) and PTD + ED high load with baghouse bypass ($n = 4$).

One and two asterisks indicate significance at a 0.05 and 0.01 probability level, respectively. Note that no distinction is made between number $N$ and mass $m$ for the separated stages because the conversion from number to mass is proportional to the size of the particles in that stage cubed, which is a constant within one stage. However, this not the case for the size fractions because these are also weighed by the particle size ranges.

No statistically significant correlation was found between DR and $N_{\text{tot}}$ or between DR and $m_{\text{tot}}$ with both dilution systems. Also, no significant correlation was observed between DR and the size fractions with $D_p > 0.01 \mu m$ which is in line with what Boman et al. (2005) concluded: the dilution ratio has no statistically significant influence on the total particle concentration for
particles from wood combustion with an aerodynamic diameter between 0.03 and 10 µm. Hedberg et al. (2002) found that the size distributions from wood combustion were not severely affected by DR when measuring with a differential mobility particle sizer (DMPS) with a range of 3 to 920 nm. However, based on our measurements, stage 1 of both ELPI+’s show a very strong positive correlation with DR. During high load operation, this correlation appears to be statistically significant while at the other operation modes this correlation is not significant (due to the limited number of tests). This is in accordance with what was already visually observed in the particle number size distributions (Fig. 6-9): an increase in DR leads to an increase of the smallest particles, measured in the ELPI+ stage 1.

However, it is more likely that this correlation is the result of measurement artefacts rather than aerosol dynamics. To illustrate this, in Fig. 11 the correlation between DR and the particles in stage 1 when diluting with the PTD + ED is shown for different boiler operation modes. When applying linear regression models, rather high coefficients of determination are obtained ($R^2 > 0.98$). Extrapolation of the models shows that at a DR of 1 (i.e. no dilution) $1.7 \times 10^6$, $1.1 \times 10^6$, and $3.0 \times 10^4$ particles per cm$^3$ can be expected during respectively high load, low load and baghouse bypass. However, these values are only indicative due to the uncertainties related with extrapolation. The linear growth of the number of particles in stage 1 with DR, and therefore with the amount of dilution air, indicates that the correlation can be the result of artefacts in the
dilution air. The ELPI+ is electrical zeroed with air that is cleaned by passing through an internal HEPA filter. The dilution air is cleaned by, among other things, passing through a HEPA filter in the air cleaning unit. A higher particle collection efficiency of the HEPA filter in the ELPI+ for the particles $D_p < 0.01 \, \mu m$ can cause a very strong correlation between DR and the particles measured in stage 1. When the flue gases are diluted before measuring, the internal HEPA filter of the ELPI+ and the HEPA filter for cleaning the dilution air should have the same collection efficiency. When this condition can not be guaranteed, it is recommended to perform the ELPI+ zeroing with the dilution air instead of using the internal HEPA filtered air. This recommendation could reduce the need to exclude stage 1 from experimental data measured with the ELPI+, an issues that often occurs. An analogous reasoning can be made with the DAD 100 but this is less pronounced because there is a relative large variation between results with the same DR. The correlation could also be explained by various other processes, both measurement artefact and aerosol dynamic related, of which some are discussed in the supplementary material, but these are believed to be less likely or unlikely.

Observing Table 3, the DAD 100 dilution system shows a strong negative correlation between DR and the particles measured in stage 8 ($r_s = -0.837$), which is significant. This correlation with the DAD 100 can be the result of particle growth by condensation and adsorption at low dilution ratio. With condensation as main gas-particle transformation, reducing DR would lead to more
condensation and thus result in a shift of the accumulation mode to the right, which was observed in Fig. 7. The occurring of condensation can be because of the fact that the DAD 100 uses lower DRs and a lower dilution air temperature (60°C) compared to the PTD + ED (150°C). The lower temperature with the DAD 100 is needed due to the temperature limit of the sample entering the ELPI+. With the PTD + ED a high temperature can be used in the PTD since the temperature is reduced in the ED. Also, the sampling probe of the DAD 100 was not heated. Condensation could occur in the probe between exiting the chimney and entering the dilution system, although this distance was limited to around 15 cm. The condensing substance can be water, since the flue gas contains about 17 vol% water, but also organic vapours are possible. Besides condensation, diluting with the DAD 100 (low DR thus less particle-free dilution air injected) allows having more frequent collisions between particles leading to more agglomeration and coagulation resulting in less ‘small’ particles and more ‘large’ particles with decreasing DR. It is rather unlikely that only one type of process (condensation or collision) occurs, but rather a combination of processes will happen in the dilution systems. However, at these concentrations the coagulation rate is rather low and are of minor importance to consider, although coagulation is an ever-present phenomenon for aerosols (Hinds, 2011; Wiinikka, 2005; Wiinikka and Gebart, 2004).
From Table 3, it can be observed that there is a strong negative correlation between DR and GMD for both the PTD + ED \((r_s = -0.986)\) and the DAD 100 \((r_s = -0.896)\) which is the result of more ‘small’ particles and less ‘large’ particles with increasing dilution. This is a consequence of the correlation between DR and stage 1.

The most appropriate DR strongly depends on the focus of the measurements. If a long measurement is required, a high DR is desirable to delay the overloading of the impactor. However, if a high measuring accuracy is required, a low DR is more desirable since the errors of measurements scale with DR. The suitable DR is thus a trade-off between the two. For this specific measurement setup and during high load without bypass, the compromise could result in a modest DR of about 20. However, to achieve the same durability for the measuring device during high load with bypass of the baghouse filters, the DR should be more than a factor 50 higher. A discussion about the influence of DR on the sampling time until the overloading of the impactor can be found in the supplementary material.

In Fig. S6 the mutual relations between \(N_{tot}, m_{tot}\), GMD and DR are visually represented. With the DAD 100 dilution system there is a relative wide spread between \(N_{tot}\) and GMD within the same operation mode compared with the PTD + ED dilution system.

\textit{Baghouse collection efficiency}
In Fig. 12 the average baghouse collection efficiency per particle size with both dilution systems is shown and is calculated with the average number of particles in each stage during high load without bypass and with the average number of particles in each stage during bypass of the baghouse. Note that the impactors of both ELPI+’s were overloaded during the baghouse bypass tests and therefore the number of particles during these tests are possibly underestimated. With the PTD + ED dilution system, the collection efficiency is higher than 98.5% for particles with $D_p > 0.05 \mu m$ whereas with the DAD 100 dilution system, the collection efficiency is higher than 97.9%. For particles with $D_p < 0.05 \mu m$ the collection efficiency decreases drastically and even reaches zero with the DAD 100 dilution system as there were less particles measured in stage 2 and 3 during the baghouse bypass than during high load without bypass. This lower collection efficiency for the smallest size fractions might be the reason why the smallest size fractions suggest the presence of a nuclei mode in the particle number size distribution.

CONCLUSIONS

The particulate emissions from a medium-scale biomass boiler at different operation modes were measured with two ELPI+’s simultaneously using two different compact partial flow dilution systems. The following conclusions can be made regarding the boiler operation mode, impactor greasing, dynamic behavior of impactors, impactor overloading and dilution conditions.
The peak of the accumulation mode shifts to a higher particle size when the boiler load decreases. For particles with $D_p > 0.05 \, \mu m$, the baghouse filters have a particle collection efficiency above 98.5% and 97.9% when using the PTD + ED and the DAD 100 dilution system respectively, but this efficiency drastically decreases when the particle size reduces.

When no grease is applied on the ELPI+ collection substrates, the concentration in stage 1 is overestimated and the concentrations in stage 3 and 4 are underestimated due to the increased bouncing effect. Hereby, the total particle number concentration can be 4 times higher and the concentration in stage 1 can be 10 times higher.

If the ELPI+ impactor is not overloaded, the particle concentrations in fractions $N_{<0.01}$ and $N_{0.01-0.1}$ reduce over time due to the effect of prematurely measuring particles in the higher stages. When starting a new measurement with a clean impactor, these fractions almost immediately decrease over time and fraction $N_{0.1-1}$ slightly increases over time due to this increasing effect. It is recommended to take this dynamic behavior of impactors into account when designing experiments and to relate the particle load on the impactors with the measured concentrations when interpreting the results.

If the ELPI+ impactor is overloaded, piles of particles collected on the impactor plates will change the cut-off diameters of the impactors. This change in cut-off diameter will shift the peak of the accumulation mode towards higher particle sizes and will result in an underestimation of
the concentrations in fractions \( N_{0.1-1} \) and \( N_{0.01-0.1} \). In addition, the saturated substrates can cause the bouncing effect to increase and thereby overestimating the concentration of fraction \( N_{<0.01} \). To postpone the effects of impactor overloading and prematurely measuring particles, oil-soaked sintered impactor plates can be used in the ELPI+.

For both dilution systems, no correlation was found with DR between \( N_{\text{tot}} \), \( m_{\text{tot}} \) or the size fractions with \( D_p > 0.01 \mu m \). However, a very strong positive correlation was found between DR and \( N_{<0.01} \) with both dilution systems. This correlation is likely the result of measurement artefacts. A difference in HEPA filtered air between the ELPI+ zeroing and the dilution air induced these measurement artefacts. It is therefore recommended to either perform the zeroing with the dilution air or either to ensure the same collection efficiency of the used HEPA filters.

Furthermore, with the DAD 100, a negative correlation was found between DR and stage 8 and it was observed that the peak of the accumulation mode shifted to higher particle sizes as DR decreases. This can be explained by the possibility that condensation was promoted with decreasing DR. The occurrence of condensation can be the result of the low DR, the low dilution air temperature and the use of an unheated probe. Both dilution systems aim to achieve a representative particle number size distribution measurement of the particulate emissions in the chimney. Provided that no measurement artefacts bias the measurements, the two stage dilution
system is preferred over one stage since with the one stage system condensation can occur. A
dilution ratio of 20 is proposed as the recommended value for this measuring setup.

SUPPLEMENTARY MATERIAL

Supplementary material can be found in the online version at www.aaqr.org.

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Table 1. Sampling conditions and particulate emissions (± standard deviation) from measurements.

<table>
<thead>
<tr>
<th>Test</th>
<th>Load</th>
<th>N_{tot}^{PTD+ED} (## cm^{-3})</th>
<th>GMD (nm)</th>
<th>N_{tot}^{DAD 100} (## cm^{-3})</th>
<th>GMD (nm)</th>
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<td>S1.1</td>
<td>HL</td>
<td>4.4  3.1 13.7±0.9 1.32 10^{5}±1.7 10^{5}</td>
<td>38±4.9</td>
<td>10.0 1.38 10^{5}±1.2 10^{5}</td>
<td>33±3.5</td>
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<td>4.0 4.50 10^{5}±3.9 10^{4}</td>
<td>133±18</td>
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<td>S2.1</td>
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<td>10.0 7.63 10^{5}±1.9 10^{5}</td>
<td>62±18</td>
</tr>
<tr>
<td>S2.2</td>
<td>HL^{b,d}</td>
<td>7.6  2.7 20.9±2.0 1.79 10^{5}±9.2 10^{4}</td>
<td>22±1.4</td>
<td>10.0 5.53 10^{5}±8.7 10^{4}</td>
<td>76±16</td>
</tr>
<tr>
<td>S2.3</td>
<td>HL^{c}</td>
<td>7.6  2.7 20.8±1.3 1.11 10^{5}±8.4 10^{4}</td>
<td>37±4.4</td>
<td>10.0 4.43 10^{5}±2.6 10^{4}</td>
<td>100±8.0</td>
</tr>
<tr>
<td>S2.4</td>
<td>HL BB^{e}</td>
<td>7.7  2.7 21.2±1.7 7.46 10^{5}±6.4 10^{5}</td>
<td>144±6.9</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
<tr>
<td>S2.5</td>
<td>HL BB^{f}</td>
<td>17.4 2.7 46.6±6.3 6.96 10^{5}±1.1 10^{5}</td>
<td>131±5.3</td>
<td>10.0 5.08 10^{5}±7.8 10^{5}</td>
<td>162±17</td>
</tr>
<tr>
<td>S2.5a</td>
<td>HL BB^{f}</td>
<td>10.6 2.7 28.8±1.9 6.51 10^{5}±6.4 10^{5}</td>
<td>145±5.1</td>
<td>10.0 4.13 10^{5}±1.1 10^{5}</td>
<td>176±2.5</td>
</tr>
<tr>
<td>S2.5b</td>
<td>HL BB^{f}</td>
<td>5.6  2.8 15.4±0.8 5.58 10^{5}±3.6 10^{5}</td>
<td>174±5.8</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
<tr>
<td>S2.6</td>
<td>LL^{g}</td>
<td>11.5 2.8 32.6±4.9 2.43 10^{5}±1.1 10^{5}</td>
<td>14±0.86</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
<tr>
<td>S2.7</td>
<td>LL^{g}</td>
<td>8.4  3.2 26.9±1.6 2.05 10^{5}±7.9 10^{4}</td>
<td>15±0.50</td>
<td>5.0 3.07 10^{5}±1.1 10^{4}</td>
<td>170±16</td>
</tr>
<tr>
<td>S2.8</td>
<td>LL^{e}</td>
<td>7.1  3.3 23.2±1.6 1.85 10^{5}±5.9 10^{4}</td>
<td>15±0.48</td>
<td>10.0 4.94 10^{5}±9.1 10^{4}</td>
<td>88±29</td>
</tr>
<tr>
<td>S2.9</td>
<td>LL^{e}</td>
<td>2.8  3.6 9.9±0.7 9.24 10^{5}±3.4 10^{4}</td>
<td>22±0.84</td>
<td>10.0 3.81 10^{5}±3.9 10^{4}</td>
<td>122±28</td>
</tr>
<tr>
<td>S2.10</td>
<td>LL^{g}</td>
<td>12.2 2.8 34.6±4.6 2.85 10^{5}±1.2 10^{5}</td>
<td>13±0.55</td>
<td>5.0 4.38 10^{5}±9.2 10^{4}</td>
<td>93±30</td>
</tr>
</tbody>
</table>

HL: high load, HL BB: high load baghouse bypass, LL: low load. n.m.: no measurement was performed. \( DR_{PTD} \): dilution ratio of the porous tube diluter; \( DR_{ED} \): dilution ratio of the ejector diluter; \( DR_{tot} \): total dilution ratio (\( = DR_{PTD} \times DR_{ED} \)).

\( N_{tot} \): total number of particles, GMD: geometric mean particle diameter.

\( ^{a} \) interchange ELPI+.

\( ^{b} \) data from PTD + ED without grease.

\( ^{c} \) data from PTD + ED rejected due to too short sampling time.

\( ^{d} \) data from DAD 100 rejected due to unstable ELPI+ pressure.

\( ^{e} \) overloading of ELPI+ impactor with PTD + ED.

\( ^{f} \) overloading of ELPI+ impactor with DAD 100.

\( ^{g} \) data from DAD 100 rejected due to too short sampling time.
Table 2. Gas concentrations (± standard deviation) measured with FTIR, Horiba, FID gas analyzers. All concentrations are dry based (except for H2O) and are corrected to a 13 vol% O2 concentration (except for H2O and O2).

<table>
<thead>
<tr>
<th>Test</th>
<th>Load</th>
<th>H2O (vol%)</th>
<th>O2 (vol%)</th>
<th>CO2 (vol%)</th>
<th>CO (mg Nm⁻³)</th>
<th>NOX (mg Nm⁻³)</th>
<th>NO (mg Nm⁻³)</th>
<th>NO2 (mg Nm⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1.1</td>
<td>HL</td>
<td>16.8±0.8</td>
<td>6.9±0.8</td>
<td>7.4±0.3</td>
<td>20.9±14.3</td>
<td>127.9±5.4</td>
<td>127.6±5.4</td>
<td>0.3±0.3</td>
</tr>
<tr>
<td>S1.2</td>
<td>HL</td>
<td>17.0±0.7</td>
<td>6.9±0.6</td>
<td>7.5±0.2</td>
<td>16.2±12.9</td>
<td>129.5±5.6</td>
<td>129.4±5.7</td>
<td>0.1±0.2</td>
</tr>
<tr>
<td>S1.3</td>
<td>HL</td>
<td>16.9±0.7</td>
<td>7.1±0.7</td>
<td>7.5±0.3</td>
<td>22.9±22.6</td>
<td>129.4±6.9</td>
<td>128.9±6.9</td>
<td>0.5±0.5</td>
</tr>
<tr>
<td>S1.4</td>
<td>HL</td>
<td>16.9±0.8</td>
<td>7.1±0.8</td>
<td>7.5±0.4</td>
<td>19.5±13.9</td>
<td>129.0±6.5</td>
<td>128.4±6.4</td>
<td>0.6±0.5</td>
</tr>
<tr>
<td>S1.5</td>
<td>HL</td>
<td>16.8±0.8</td>
<td>7.1±0.9</td>
<td>7.5±0.4</td>
<td>15.8±11.5</td>
<td>130.0±5.8</td>
<td>129.6±5.7</td>
<td>0.4±0.4</td>
</tr>
<tr>
<td>S2.1</td>
<td>HL</td>
<td>16.7±0.6</td>
<td>7.1±0.6</td>
<td>7.3±0.3</td>
<td>24.8±11.2</td>
<td>125.1±6.8</td>
<td>122.8±6.4</td>
<td>6.4±1.4</td>
</tr>
<tr>
<td>S2.2</td>
<td>HL</td>
<td>16.8±0.7</td>
<td>7.5±0.5</td>
<td>7.6±0.5</td>
<td>21.3±12.7</td>
<td>133.8±8.8</td>
<td>130.5±7.9</td>
<td>7.9±1.9</td>
</tr>
<tr>
<td>S2.3</td>
<td>HL</td>
<td>16.9±0.6</td>
<td>6.9±0.7</td>
<td>7.4±0.3</td>
<td>21.9±7.1</td>
<td>125.8±6.3</td>
<td>122.1±6.9</td>
<td>6.9±2.2</td>
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<tr>
<td>S2.4</td>
<td>HL BB</td>
<td>16.9±0.4</td>
<td>6.9±0.5</td>
<td>7.3±0.2</td>
<td>29.5±12.8</td>
<td>124.8±4.9</td>
<td>121.2±5.2</td>
<td>5.2±2.1</td>
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<tr>
<td>S2.5</td>
<td>HL BB</td>
<td>17.1±1.1</td>
<td>6.6±0.6</td>
<td>7.1±0.6</td>
<td>23.2±13.4</td>
<td>122.6±11.6</td>
<td>119.0±13.0</td>
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<td>HL BB</td>
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<td>6.9±0.6</td>
<td>7.3±0.2</td>
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<td>123.2±3.8</td>
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<td>HL BB</td>
<td>17.1±0.6</td>
<td>7.0±0.6</td>
<td>7.3±0.2</td>
<td>43.6±37.6</td>
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<td>124.6±6.0</td>
<td>6.0±1.4</td>
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<td>LL</td>
<td>15.1±0.6</td>
<td>7.9±0.5</td>
<td>7.2±0.3</td>
<td>55.2±37.3</td>
<td>107.7±15.5</td>
<td>87.2±20.0</td>
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<td>LL</td>
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<td>7.8±0.4</td>
<td>7.3±0.3</td>
<td>120.3±29.8</td>
<td>75.3±10.0</td>
<td>58.3±6.9</td>
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<td>LL</td>
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<td>7.9±0.6</td>
<td>7.2±0.4</td>
<td>214.5±67.7</td>
<td>63.6±5.7</td>
<td>45.5±3.8</td>
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<tr>
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<td>LL</td>
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<td>7.9±0.9</td>
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<td>S2.10</td>
<td>HL</td>
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<td>27.8±39.7</td>
<td>108.8±16.9</td>
<td>97.7±22.6</td>
<td>22.6±6.6</td>
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<table>
<thead>
<tr>
<th>Test</th>
<th>Load</th>
<th>O2 (vol%)</th>
<th>CO2 (vol%)</th>
<th>CO (mg Nm⁻³)</th>
<th>NOX (mg Nm⁻³)</th>
<th>NO (mg Nm⁻³)</th>
<th>FID TOC (mg C Nm⁻³)</th>
</tr>
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<tbody>
<tr>
<td>S1.1</td>
<td>HL</td>
<td>6.9±0.6</td>
<td>7.8±0.3</td>
<td>28.6±14.8</td>
<td>163.6±7.6</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
<tr>
<td>S1.2</td>
<td>HL</td>
<td>6.6±0.5</td>
<td>8.0±0.3</td>
<td>22.4±11.5</td>
<td>167.8±7.3</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
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<td>S1.3</td>
<td>HL</td>
<td>6.7±0.6</td>
<td>8.0±0.3</td>
<td>26.1±19.9</td>
<td>171.1±9.5</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
<tr>
<td>S1.4</td>
<td>HL</td>
<td>6.8±0.7</td>
<td>7.9±0.4</td>
<td>22.2±12.4</td>
<td>173.6±9.2</td>
<td>n.m.</td>
<td>n.m.</td>
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<tr>
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<td>HL</td>
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<td>7.9±0.4</td>
<td>19.4±9.9</td>
<td>175.0±9.4</td>
<td>n.m.</td>
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<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
</tr>
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<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>3.1±1.5</td>
<td>n.m.</td>
</tr>
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<td>HL</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>2.5±1.7</td>
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<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>3.8±1.9</td>
<td>n.m.</td>
</tr>
<tr>
<td>S2.5</td>
<td>HL BB</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>n.m.</td>
<td>3.5±1.4</td>
<td>n.m.</td>
</tr>
<tr>
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<td>HL BB</td>
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<td>n.m.</td>
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<td>n.m.</td>
<td>3.5±1.4</td>
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</tr>
<tr>
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<td>n.m.</td>
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<td>7.7±0.3</td>
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<td>158.0±19.3</td>
<td>1.5±1.0</td>
<td>n.m.</td>
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<tr>
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<td>n.m.</td>
</tr>
<tr>
<td>S2.8</td>
<td>LL</td>
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<td>7.8±0.4</td>
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<td>1.2±0.7</td>
<td>n.m.</td>
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<tr>
<td>S2.9</td>
<td>LL</td>
<td>7.7±0.7</td>
<td>7.8±0.4</td>
<td>148.5±35.6</td>
<td>77.2±16.0</td>
<td>0.6±0.4</td>
<td>n.m.</td>
</tr>
<tr>
<td>S2.10</td>
<td>HL</td>
<td>6.5±1.0</td>
<td>7.9±0.5</td>
<td>37.6±38.4</td>
<td>144.0±29.8</td>
<td>0.3±0.3</td>
<td>n.m.</td>
</tr>
</tbody>
</table>

HL: high load, HL BB: high load baghouse bypass, LL: low load, n.m.: no measurement was performed.

TOC concentrations are close to the limit of quantification.
Table 3. Spearman correlation coefficient $r_s$ between DR and particle number fractions, particle mass fractions, each stage separately and GMD.

<table>
<thead>
<tr>
<th></th>
<th>PTD + ED</th>
<th>DAD 100</th>
<th>PTD + ED</th>
<th>PTD + ED</th>
</tr>
</thead>
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<tr>
<td></td>
<td>HL</td>
<td>HL</td>
<td>LL</td>
<td>HL BB</td>
</tr>
<tr>
<td></td>
<td>$N  $</td>
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<td>$N  $</td>
<td>$m$</td>
</tr>
<tr>
<td>Total</td>
<td>0.543</td>
<td>0.657</td>
<td>0.777</td>
<td>-0.120</td>
</tr>
<tr>
<td>$1 \mu m &lt; D_p &lt; 10 \mu m$</td>
<td>0.600</td>
<td>0.657</td>
<td>0.060</td>
<td>-0.060</td>
</tr>
<tr>
<td>$0.1 \mu m &lt; D_p &lt; 1 \mu m$</td>
<td>0.029</td>
<td>0.486</td>
<td>0.239</td>
<td>-0.657</td>
</tr>
<tr>
<td>$0.01 \mu m &lt; D_p &lt; 0.1 \mu m$</td>
<td>-0.371</td>
<td>-0.086</td>
<td>0.717</td>
<td>0.777</td>
</tr>
<tr>
<td>$D_p &lt; 0.01 \mu m$</td>
<td>1.00**</td>
<td>1.00**</td>
<td>0.896*</td>
<td>0.896*</td>
</tr>
<tr>
<td>Stage 1</td>
<td>1.00**</td>
<td>0.896*</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>Stage 2</td>
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<td>0.949</td>
<td>1.00</td>
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<tr>
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<td>0.418</td>
<td>0.949</td>
<td>0.800</td>
</tr>
<tr>
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<td>0.777</td>
<td>0.800</td>
<td>-0.400</td>
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<td>-0.400</td>
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<td>-0.400</td>
</tr>
<tr>
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<td>-0.800</td>
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<td>Stage 10</td>
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<td>-0.200</td>
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<td>0.400</td>
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<td>Stage 13</td>
<td>0.600</td>
<td>0.00</td>
<td>-0.400</td>
<td>0.800</td>
</tr>
<tr>
<td>Stage 14</td>
<td>-0.135</td>
<td>0.422</td>
<td>1.00</td>
<td>0.800</td>
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<tr>
<td>GMD</td>
<td>-0.986**</td>
<td>-0.896*</td>
<td>-0.949</td>
<td>-0.800</td>
</tr>
</tbody>
</table>

HL: high load, LL: low load, HL BB: high load baghouse bypassed.

* Significant at the 0.05 probability level.
** Significant at the 0.01 probability level.
Figure Captions

**Fig. 1.** Sampling setup for the PTD + ED dilution system, the DAD 100 dilution system and the flue gas analyzers.

**Fig. 2.** Time series of emissions during S1 with boiler operating at high load. (a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.

**Fig. 3.** Time series of emissions during S2.1 to S2.3 and S2.10 with boiler operating at high load. (a) Particulate emissions when diluting with the PTD + ED dilution system. N₁₀₀ and N₀.₀₁ during test S2.1 (i.e. no grease test) are drawn relative to the grey scale since they are of a different order of magnitude. The impactor of ELPI+ A was overloaded during test S2.10. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.

**Fig. 4.** Time series of emissions during S2.4 to S2.5b with boiler operating at high load with the baghouse bypassed. (a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.
Fig. 5. Time series of emissions during S2.6 to S2.9 with boiler operating at low load.

(a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.

Fig. 6. Particle number size distributions at high load with the PTD + ED dilution system.

i.c.: ELPI+ interchange, n.g.: no grease, o.l.: impactor overload.

Fig. 7. Particle number size distributions at high load with the DAD 100 dilution system.

i.c.: ELPI+ interchange, u.p.: unstable ELPI+ pressure.

Fig. 8. Particle number size distributions at low load. o.l.: impactor overload.

Fig. 9. Particle number size distributions at high load with the baghouse bypassed. o.l.: impactor overload.

Fig. 10. The correlation of (a) total particle number \( N_{\text{tot}} \), (b) total particle mass \( m_{\text{tot}} \) and (c) geometric mean diameter GMD between the PTD + ED and DAD 100 dilution systems.

Fig. 11. Correlation between DR and particles measured in stage 1 when diluting with PTD + ED.

HL: high load, LL: low load, HL BB: high load with baghouse bypassed.

Fig. 12. Average baghouse collection efficiency per particle size.
Fig. 1. Sampling setup for the PTD + ED dilution system, the DAD 100 dilution system and the flue gas analyzers.
Fig. 2. Time series of emissions during S1 with boiler operating at high load. (a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O$_2$.
Fig. 3. Time series of emissions during S2.1 to S2.3 and S2.10 with boiler operating at high load. (a) Particulate emissions when diluting with the PTD + ED dilution system. $N_{\text{tot}}$ and $N_{<0.01}$ during test S2.1 (i.e. no grease test) are drawn relative to the grey scale since they are of a different order of magnitude. The impactor of ELPI+ A was overloaded during test S2.10. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O$_2$.
Fig. 4. Time series of emissions during S2.4 to S2.5b with boiler operating at high load with the baghouse bypassed. (a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.
Fig. 5. Time series of emissions during S2.6 to S2.9 with boiler operating at low load.

(a) Particulate emissions when diluting with the PTD + ED dilution system. (b) Particulate emissions when diluting with the DAD 100 dilution system. (c) Gaseous emissions measured with FTIR. Gas concentrations are dry based and corrected to 13 vol% O₂.
Fig. 6. Particle number size distributions at high load with the PTD + ED dilution system.

i.c.: ELPI+ interchange, n.g.: no grease, o.l.: impactor overload.
**Fig. 7.** Particle number size distributions at high load with the DAD 100 dilution system.

d.: $d N/d \log D_p (\#/cm^3)$

i.c.: ELPI+ interchange, u.p.: unstable ELPI+ pressure.
Fig. 8. Particle number size distributions at low load. o.l.: impactor overload.
Fig. 9. Particle number size distributions at high load with the baghouse bypassed. o.l.: impactor overload.
Fig. 10. The correlation of (a) total particle number $N_{tot}$, (b) total particle mass $m_{tot}$ and (c) geometric mean diameter GMD between the PTD + ED and DAD 100 dilution systems.
Fig. 11. Correlation between DR and particles measured in stage 1 when diluting with PTD + ED.

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