# Supplementary Material

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

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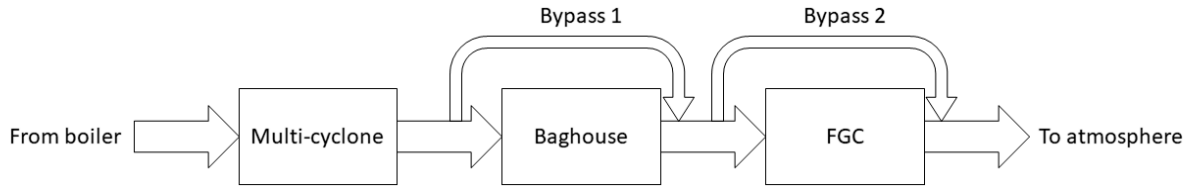
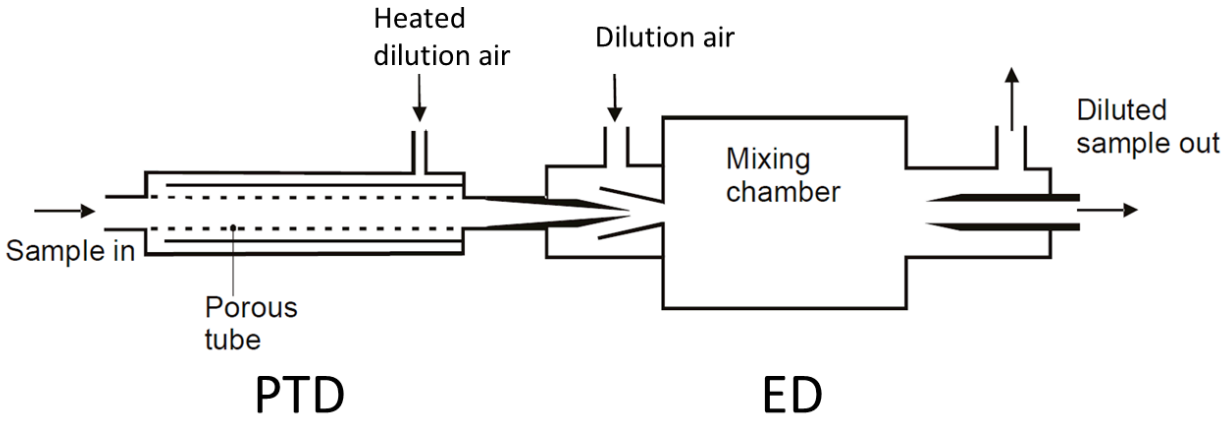



**Fig. S1.** Schematic representation of the pathway of the flue gas from boiler to atmosphere. FGC: flue gas condenser.

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24 **Fig. S2.** Two stage dilution system: porous tube diluter (PTD) in combination with ejector

25 diluter (ED). Figure based on Jokiniemi *et al.* (2008).

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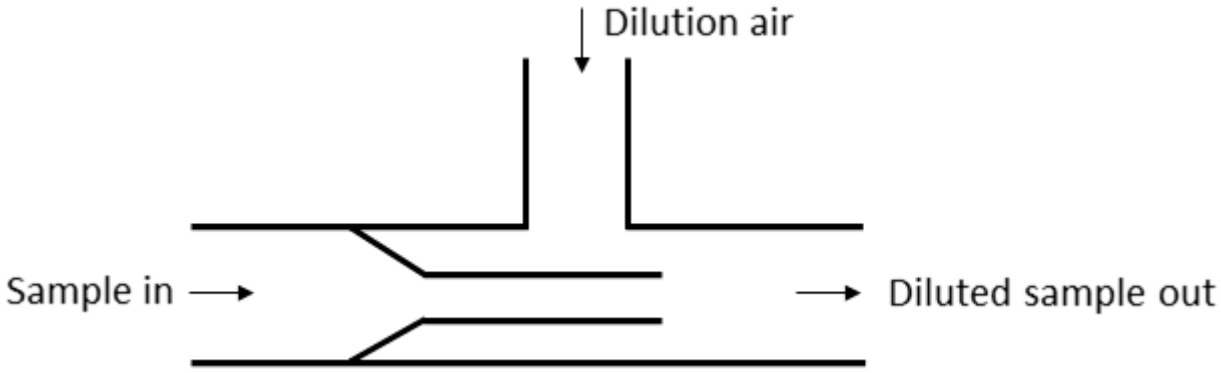


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28 **Fig. S3.** One stage dilution system: Dekati axial diluter (DAD 100). Figure based

29 on Dekati (2010).

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***Particle mass size distributions***

Analogous to the number distribution, the particle mass size distribution expresses the mass of the particles within a size interval divided by the logarithmic range of the size interval (d$m$ / d log $D_p$ on vertical axis) at the geometric mean of each size interval ($D_p$ on horizontal axis). This distribution is not measured directly but rather calculated with the ELPI+ results using the estimated particle density of $\rho$ = 2.0 g cm$^{-3}$ and assuming spherical particles. Although the particle number size distribution is more appropriate to relate particulate emissions to its health impact, the particle mass size distribution can also be useful to compare both dilution systems, especially for the particle fractions with the largest particles.

However, the sampling was not isokinetic since the flue gas velocity in the chimney is higher than the sampling velocity so caution is necessary for drawing conclusions for coarse particles. The maximum sampling error that occurs due to anisokinetic sampling is estimated to be lower than 0.5% for $D_p$ < 0.1 µm when using the method described by Belyaev and Levin (1974). For particles of 1 µm this error is estimated to be 2.2% at the lowest DR but reaches 22% at the highest DR. For particles of 10 µm the error is estimated to be 95% at the lowest DR and almost a tenfold of this for the highest DR. Furthermore, Pagels *et al.*, (2005) found that the original ELPI overestimates the coarse mode which is likely due to fine particle losses (i.e. secondary particle collection) which can result in non-negligible mass in the upper stages. They concluded that the ELPI can give reliable high time- and size-resolved mass size distributions up to 3 µm. Järvinen

*et al.* (2014) found that the new ELPI+ had a similar secondary collection of nanoparticles than the ELPI for stages with the largest cut-off diameters. In addition, almost all coarse particles are trapped by the multi-cyclone as the particles with $D_p > 2.5$ µm only represent less than 0.04% of the total number of particles in this study. For these reasons, only particles with $D_p < 3$ µm are considered here in the particle mass size distribution, namely stages 1 to 11.

In Fig. S4 and S5 the particle mass size distributions with the two dilution systems at various tests are presented. With both dilution systems and during all operation modes, an accumulation mode can be distinguished between 0.1 and 1 µm, which is in line with previous studies regarding the particle mass size distribution of wood combustion (Lamberg *et al.*, 2011; Tissari *et al.*, 2008b; Krugly *et al.*, 2014; Hays *et al.*, 2003; Wierzbicka *et al.*, 2005; Lillieblad *et al.*, 2004). During some tests a second mode can be distinguished in the coarse fraction of the particle mass size distributions which was also observed in previous studies (Lamberg *et al.*, 2011; Tissari *et al.*, 2008a,b). However, by only considering particles with $D_p < 3$ µm this mode is not fully covered. Note that the mass of particles in the size fractions with the smallest particles is negligibly small since the mass varies with the diameter of the particles cubed. In addition, due to the same reason, a small number of particles in the largest size fractions result in a high particle mass concentration.

In Fig. S4a the average of the particle mass size distributions for the two dilution systems during high load and low load are presented. The high load distributions are the averages of tests S1.1 to S1.5 for both dilution systems. The low load distribution with the PTD + ED is the average of tests S2.6 to S2.9 that were carried out with the overloaded ELPI+ impactor. The low load distribution with the DAD 100 is the average of tests S2.8 and S2.9. With the DAD 100 both distributions are quite similar although the low load distribution appears to be slightly lower. With the PTD + ED high load distribution, the accumulation peak appears to be higher compared to the DAD 100. Also, stage 9 is higher and stage 8 is lower than expected. By studying the particle mass size distributions of the individual tests, it was observed that as time goes on, the particle concentration in stage 9 increases and the concentration in stage 8 decreases. This is probably due to a change in cut-off diameter of stage 9 resulting in the particles of stage 8 being prematurely measured in stage 9. With the PTD + ED low load distribution, the accumulation mode is higher and appears to be slightly shifted towards larger particles.

In Fig. S4b the particle mass size distribution of test S2.1 with both dilution systems is drawn. Although this is the same operation mode as the high load in Fig. S4a, there are differences. During test S2.1 with the DAD 100, the peak of the accumulation mode is less pronounced and a small increase in the coarse fraction can be observed. With the PTD + ED no grease was used

84 during this test. The accumulation mode appears to be higher and an increase in the coarse
85 fraction can be observed.
86 In Fig. S4c the particle mass size distribution of test S2.3 with both dilution systems is
87 presented. With the DAD 100 there was an unstable pressure in the ELPI+, but the distribution
88 appears to be similar to the distribution of tests S2.1. With the PTD + ED grease was used during
89 this tests but the distribution is similar to the distribution of tests S2.1 without grease. Only
90 stage 11 during S2.3 appears to be higher.
91 In Fig. S4d the particles mass size distribution of test S2.10 is shown for both dilution systems.
92 The test with the PTD + ED was measured with an overloaded ELPI+ impactor. The height of the
93 accumulation mode is similar as the one of the average high load tests in Fig. S4a. With the
94 DAD 100, the peak is more toward larger particles.
95 In Fig. S5 the particle mass size distributions are shown of the tests during high load with the
96 baghouse bypassed. During these tests, both ELPI+ impactors were overloaded with particles.
97 The peak of the accumulation mode appears to be lower for the DAD 100 compared with the
98 PTD + ED. With the PTD + ED, all tests show a shoulder in the coarse fraction while this is not
99 the case with the DAD 100.
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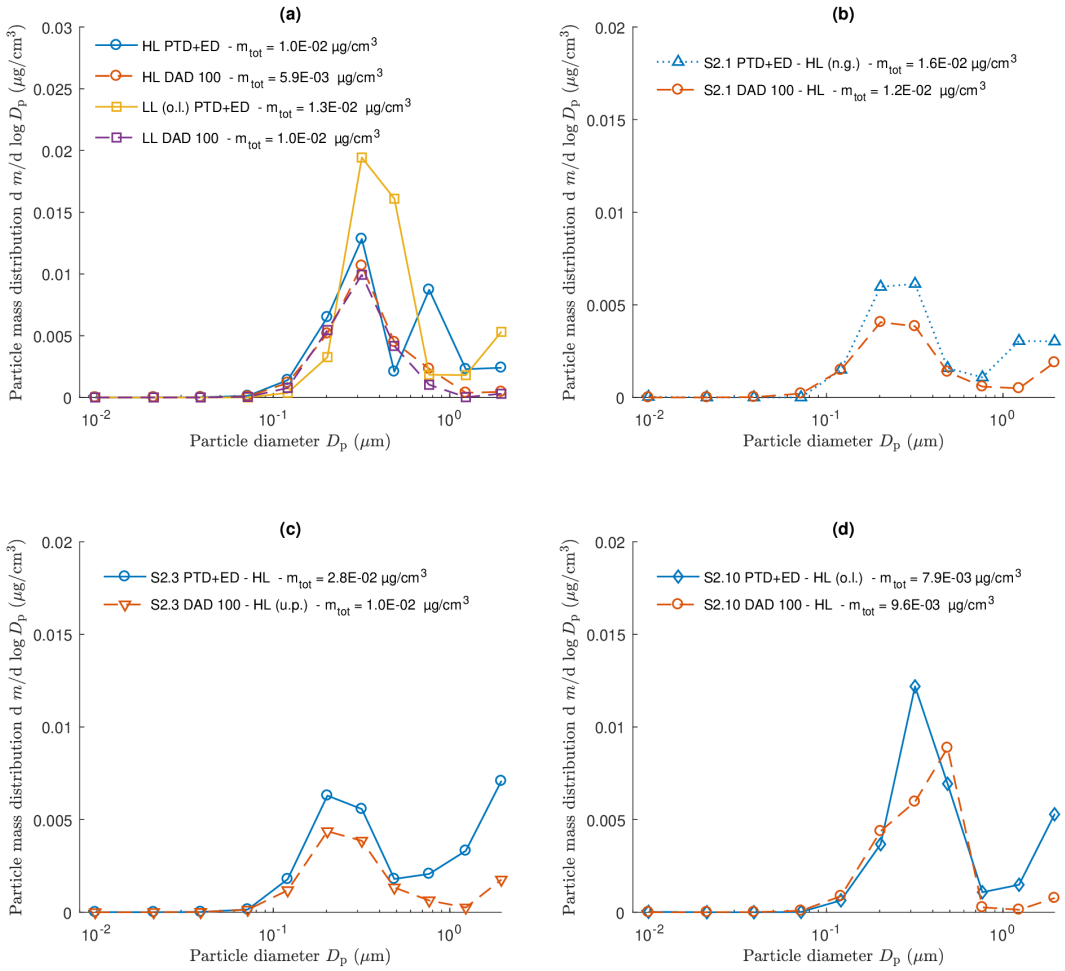


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103 **Fig. S4.** Particle mass size distributions during high load (HL) and low load (LL) with both

104 dilution systems. (a) HL are the average of tests S1.1 to S1.5. LL with PTD + ED is the average

105 of tests S2.6 to S2.9 and measured with the ELPI+ impactor overloaded (o.l.). LL with DAD 100

106 is the average of tests S2.8 and S2.9. (b) Simultaneous tests with no grease (n.g.) applied with

107 ELPI+ A. (c) Simultaneous tests with unstable pressure (u.p.) is ELPI+ B. (d) Simultaneous tests

108 with impactor of ELPI+ A overloaded (o.l.).

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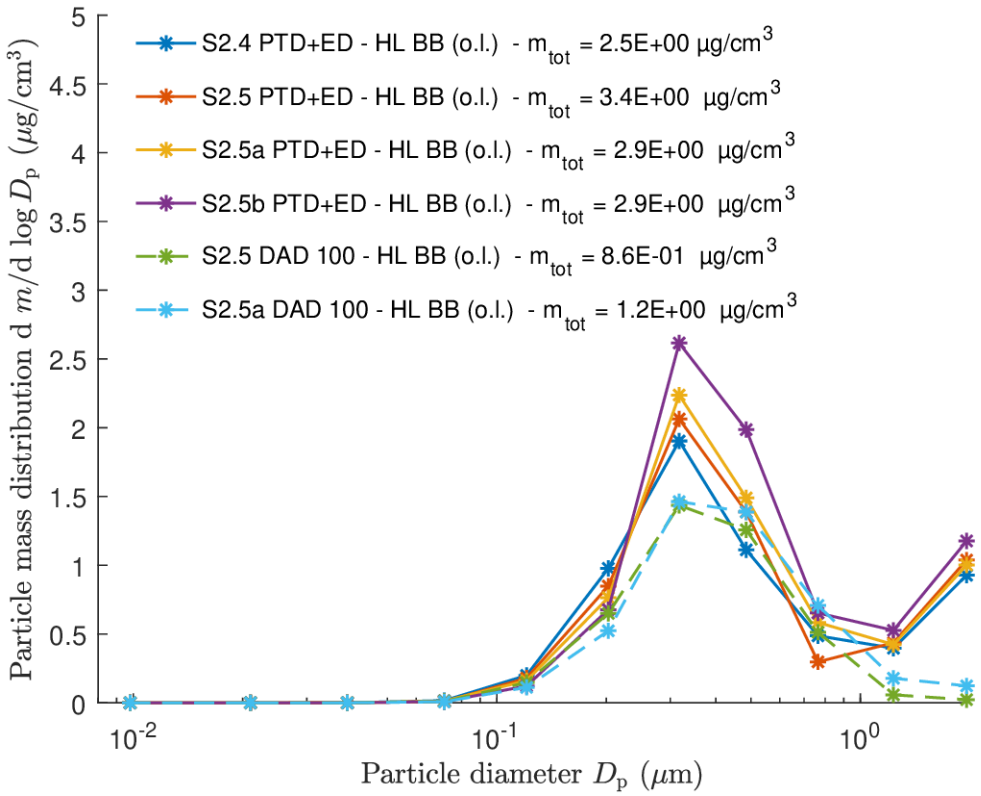


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111 **Fig. S5.** Particle mass size distributions during high load with bypass of the baghouse with both

112 dilution systems.

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***Alternative reasons for correlation between DR and particle concentration in stage 1***

The correlation could also be explained by various other processes, both measurement artefact and aerosol dynamic related, of which some will be discussed here but these are believed to be less likely or unlikely.

(i) A possible explanation is that DR was incorrectly estimated. This is however unlikely since an incorrect DR would also cause a correlation with $N_{tot}$. Moreover, the good agreement of $N_{0.1\text{-}1}$ between different tests and the different dilution systems (e.g. see Fig. 2) indicates that DR is indeed estimated correctly.

(ii) Another explanation is that the estimated particle density of 2.0 g $cm^{-3}$ would induce such a change in the size distribution causing stage 1 to correlate with DR. However, the same correlation can be found when using a density of 1.0 g $cm^{-3}$ since a change in density causes a rescaling of the distribution rather than a distortion.

(iii) Another possibility is the correction algorithm of the ELPI+ that corrects the lower stages for particle loss in the higher stages. However, investigation of the uncorrected data showed that the correlation is present even without the correction. The corrected number concentration in stage 1 is about 2.7 times the uncorrected concentration for all high load tests. This correction factor depends on the currents measured in the higher stages and is almost constant during the same operation mode. This correction factor is around 3.1 during the baghouse bypass and 2.7 during low load and changes little with the chosen particle density.

(iv) The correlation could be the result of increasing particle diffusional losses in the sampling system with decreasing DR. Diffusion losses increase with increasing tube length, decreasing particle size and decreasing flow rate (Brockmann, 2011). The tubes from the dilution systems to the ELPI+'s were approximate 1 m and did not change with DR. Smaller particles will deposit easier due to diffusion but there is no indication that particles reduce in size with decreasing DR. The total flow rate in the DAD 100 is constant and does not change with DR. The flow rate in the PTD decreases with decreasing DR which would result in an increase in diffusion losses, however, due to the construction of the PTD particle deposition is minimized. The dilution ratio of the ED is about constant and independent of the flow rate in the ED. Thus, the flow rate in the ED does not correlate with the total DR. Therefore, it is rather unlikely that the correlation between DR and the particle concentration in stage 1 is the result of increased diffusion deposition with decreasing DR.

(v) The significant correlation observed between DR and particles in stage 1, could be the result of the formation of new particles by increased nucleation with increasing DR. The dynamic behavior of the gas-particle system in the dilution process will determine the number size distribution after dilution and cooling (Lyyränen *et al.*, 2004). Research on diesel particle emissions has shown that the nuclei mode is subjected to dilution conditions (Kittelson *et al.*, 2006). It has been observed with diesel emissions that increasing dilution favors the formation of

151 particles with $D_p < 0.05$ µm (Shi and Harrison, 1999; Shi *et al.*, 2000) but also a decrease of
152 nanoparticles with increasing dilution has been reported in literature (Jaiprakash *et al.*, 2016;
153 Suresh and Johnson, 2001; Abdul-Khalek *et al.*, 1999; Lipsky and Robinson, 2006). With diesel
154 exhaust, nucleation is possible because the nucleating species (mainly sulfuric acid and
155 hydrocarbons) are generally in the vapour phase in the tailpipe, and undergo a gas-to-particle
156 conversion during dilution and cooling (Kittelson *et al.*, 2006). With biomass combustion, only a
157 limited amount of gaseous species relevant for gas-to-particle conversion are present in the flue
158 gas at 150°C. At these conditions, probably only $Na_2SO_4$, HCl and $ZnCl_2$ and less likely $PbCl_2$
159 can be expected (Jöller *et al.*, 2005). Also organic vapours can be expected in the flue gas but
160 only in low concentrations due to the high combustion efficiency. Nucleation of new particles is a
161 competing process with adsorption and condensation on pre-existing particles (Lyyränen *et al.*,
162 2004). Therefore, besides nucleation, also condensation should occur since the particles in the
163 accumulation mode provide surface to condense on. There is however no positive significant
164 correlation with DR and particle concentration in the accumulation mode (see Table 3). Moreover,
165 the opposite was observed in Fig. 7, with decreasing DR the peak of the accumulation mode
166 shifted to larger particles sizes. In addition, nucleation of new particles requires a large saturation
167 ratio (Hinds, 2011). Since with the PTD + ED the dilution occurs in two stages (i.e. first
168 decreasing the vapour pressure and then decreasing the temperature), the flue gas is not expected

169 to become supersaturated during dilution and thus nucleation of new particles in the dilution

170 system is not likely.

171

***Sampling time until impactor overload***

The sampling time until particle overload in the impactors can be estimated based on the dilution ratio and the actual measured distributions. The maximum particle load given by the ELPI+ user manual is 1 mg (Dekati, 2016). However, van Gulijk *et al.* (2001) showed that after collecting only 15 µg of particles, the relative error on the measured particle number size distribution is already 10% due to the prematurely measuring of small particles. In Table S1, the sampling time until overload is given for both maximum particle loads (i.e. $m_{max}$ = 1 mg and 15 µg). The column *accum. mode* indicates the time until at least one impactor stage of the accumulation mode reaches the maximum particle load. Analogue, the column *coarse mode* indicates the time until at least one impactor stage of the coarse mode reaches the maximum particle load. The sampling times are estimated for high boiler load without baghouse bypass (HL) and for high boiler load with baghouse bypass (HL BB) and for three different dilution ratios. The dilution ratios are the typical DR of the DAD 100 dilution system (DR = 10), the recommended DR for the PTD + ED dilution system (DR = 20) and a high DR of 1000 (by extrapolating the estimations of DR = 20).

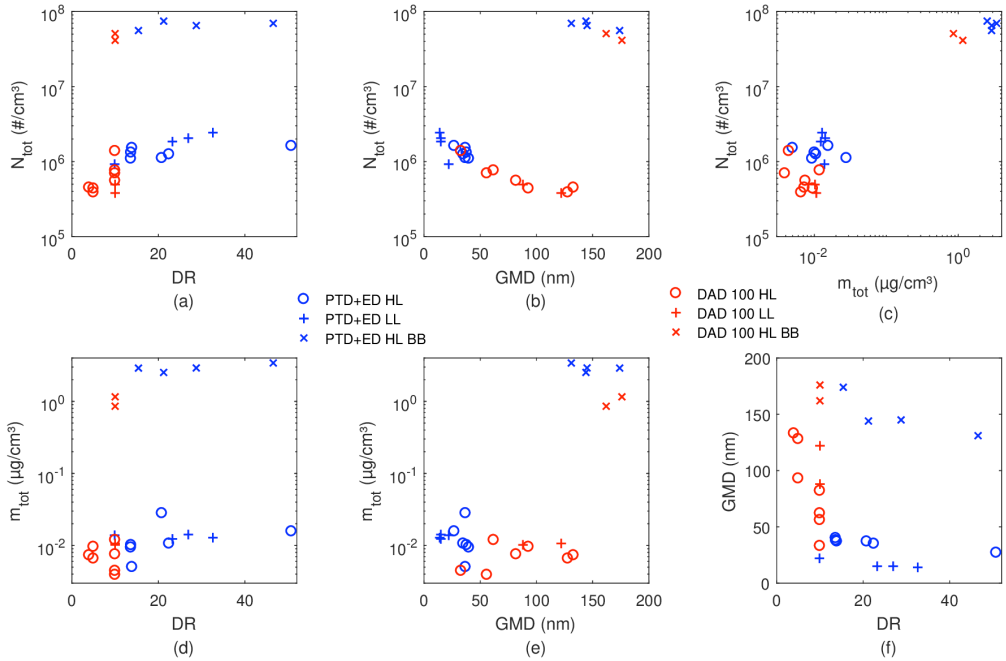
Both dilution systems can be used at least 13 h to sample emissions during HL before one of the impactor stages reaches 1 mg and needs cleaning. However, after only about 12 minutes of sampling the relative errors become larger than 10% due to the prematurely measuring of the smallest particles.

Both dilution systems can be used less than 4 minutes to sample emissions during HL BB before one of the impactor stages reaches 1 mg and needs cleaning. After less than 4 s the relative errors become larger than 10%. Using a dilution system with DR = 1000 would extend the sampling time before overload to over 2 h but after less than 2 minutes the relative error would exceed 10%. In any case, keeping the relative error below 10% by limiting the sampling time is a rather inconvenient method for carrying out measurements.

It is recommend to take this dynamic behavior of impactors into account when designing an experiment and to relate the particle load on the impactors with the obtained concentrations when interpreting the results.

**Table S1.** Sampling time until at least one impact stage is overloaded.

| Load | Dilution system | DR | $m_{max}$ = 1 mg | | $m_{max}$ = 15 µg | |
|---|---|---|---|---|---|---|
| | | | accum. mode | coarse mode | accum. mode | coarse mode |
| HL | | | | | | |
| | DAD 100 | 10 | 13.9 h | 75.3 h | 12.5 min | 67.8 min |
| | PTD + ED | 20 | 20.8 h | 13.3 h | 18.8 min | 12.0 min |
| | | 1000 | 1040 h | 667 h | 938 min | 600 min |
| HL BB | | | | | | |
| | DAD 100 | 10 | 3.7 min | 20.0 min | 3.3 s | 18.0 s |
| | PTD + ED | 20 | 6.8 min | 2.5 min | 6.1 s | 2.3 s |
| | | 1000 | 341 min | 126 min | 307 s | 113 s |

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205 **Fig. S6.** The mutual relations between total particle number $N_{tot}$, total particle mass $m_{tot}$,

206 geometric mean particle diameter GMD and dilution ratio DR. HL: high load, LL: low load, HL

207 BB: high load with baghouse bypassed.

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