Influence of soot aerosol properties on the counting efficiency of PN-PTI instruments

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8 Abstract. In this work, we investigated the influence of different types of soot aerosol on the counting efficiency 9 (CE) of instruments employed for the periodic technical inspection (PTI) of diesel vehicles. Such instruments 10 report particle number (PN) concentration. Combustion aerosols were generated by a prototype bigCAST, a 11 miniCAST 5201 BC, a miniCAST 6204 C and a miniature inverted soot generator (MISG). For comparison 12 purposes, diesel soot was generated by a Euro 5b diesel test vehicle with by-passed diesel particulate filter (DPF). 13 The size-dependent counting efficiency profile of six PN-PTI instruments was determined with each one of the 14 aforementioned test aerosols. The results showed that the type of soot aerosol affected the response of the PN-PTI 15 sensors in an individualised manner. Consequently, it was difficult to identify trends and draw conclusive results 16 about which laboratory-generated soot is the best proxy for diesel soot. Deviations in the counting efficiency 17 remained typically within 0.25 units when using laboratory-generated soot compared to Euro 5b diesel soot of 18 similar mobility diameter (~50-60 nm). Soot with a mobility diameter of ~100 nm generated by the MISG, the 19 lowest size we could achieve, resulted in most cases in similar counting efficiencies as that generated by the 20 different CAST generators at the same particle size-for most of the PN PTI instruments, showing that MISG may

$21 \qquad \text{be a satisfactory - and affordable - option for PN-PTI verification.}$

22 1 Introduction

- 23 Soot particles emitted by transport sources can have adverse health effects (Kheirbek et al., 2016; US-EPA, 2019;
- 24 WHO, 2021). To reduce particulate emissions, new procedures for the periodic technical inspection (PTI) of diesel
- 25 vehicles based on the measurement of particle number (PN) concentration have recently been established in
- Switzerland, Germany, the Netherlands and Belgium, while other countries might follow in due time (EU, 2023;
 Vasilatou et al., 2022). Portable instruments known as PN-PTI counters are used for measuring particle number
- 28 concentration (PNC) directly in the tailpipe of diesel vehicles equipped with a diesel particle filter (DPF)
- 29 (Kesselmeier and Staudt, 1999; Melas et al., 2021, 2022, 2023). When the DPF is intact, the emitted PNC is low
- 30 (typically up to a few thousand particles per cm³), whereas if the DPF is defect or tampered, PNC increases to
- 31 several hundred thousand particles per cm³ (Botero et al., 2023; Burtscher et al., 2019; Giechaskiel et al., 2022).
- 32 In terms of particle mass concentration, a functioning DPF can reduce particulate emissions by up to a factor of
- 33 150 (Ligterink, 2018) while in terms of particle number concentration a solid particle number trapping efficiency
- 34 of higher than 99 % has been reported in the literature (Frank, Adam et al., 2020). It has been shown that a small
- 35 fraction (about 10 %) of vehicles with defective DPF is responsible for up to 80-90 % of the total fleet emissions
- 36 (Burtscher et al., 2019; Kurniawan and Schmidt-Ott, 2006). The goal of PN-PTI procedures is to identify diesel

vehicles with compromised DPFs, thus ensuring that vehicles in operation maintain their performance as
 guaranteed by type-approval, without excessive degradation, throughout their lifetime (EU, 2023).

39 Although the concept of PN-PTI is simple, its implementation in practice is not as straightforward. PTI procedures

40 are not fully harmonised and, as a result, the limit values for the emitted PNC, the technical specifications of the

PN-PTI counters and the test protocol for type-examination and verification are defined at a national level (Anon,
2019; AU-Richtlinie, n.d.; PTB, 2021; UVEK, 2023; VAMV, 2018; Vasilatou et al., 2022, 2023). Differences in

43 national legislations might lead to contradicting results, e.g. the same diesel vehicle might pass the PTI check in

44 one country but fail in another one. To ensure fair implementation of regulations across Europe and avoid

- unnecessary costs which may occur for vehicle owners after a False Fail, the various PTI procedures must becompared and the differences elucidated.
- 47 PN-PTI instruments go through a type-examination procedure which may differ in each country. Among several

48 tests, type-examination includes a counting efficiency and a linearity check typically performed with combustion

49 aerosols. During their lifetime, PN-PTI instruments are checked for their linearity with polydisperse particles

50 (typically with a GMD of 70 ± 20 nm). In our previous study (Vasilatou et al., 2023), we showed that the choice

51 of test aerosol during type-examination or verification of PN-PTI instruments significantly affects the performance

52 of instruments based on diffusion charging (DC). When sodium chloride (NaCl) or carbonaceous particles from

53 spark-discharge generators were used as test aerosols, the counting efficiency of the DC-based instruments

54 changed by up to a factor of two compared to that exhibited with diesel soot. The experiments clearly showed that

- soot from laboratory-based combustion generators was the best proxy for soot emitted by diesel engines, however,
- 56 potential differences between the different combustion generators available on the market were not investigated.
- 57 In this study, we challenged six different DC-based PN-PTI instruments with polydisperse soot particles produced
- 58 by three different CAST generators (Jing AG, Switzerland), the miniature inverted soot generator (MISG,

59 Argonaut Scientific, Canada) and a Euro 5b diesel vehicle. The geometric mean diameter of the test aerosol was

60 in the range used for linearity checks of PN-PTI instruments as well as in typical size range emitted by diesel

61 engines. The scope of our study was to investigate possible differences that may arise when using different

62 combustion aerosol generators during the type-examination and verification of PN-PTI instruments as well as to

63 correlate with diesel engine emitted soot. We focused on DC-based instruments because we expect a larger impact

- 64 of the aerosol properties on their response compared to CPC-based ones (Vasilatou et al., 2023). The size-
- 65 dependent counting efficiency of the PN-PTI instruments was determined by using a condensation particle counter
- 66 (NPET 3795, TSI Inc., USA) as a reference instrument. We discuss the results in the context of the different
- 67 national legislations and make recommendations for the harmonisation of the various calibration and verification
- 68 procedures in the laboratory.

69 2 Materials and methods

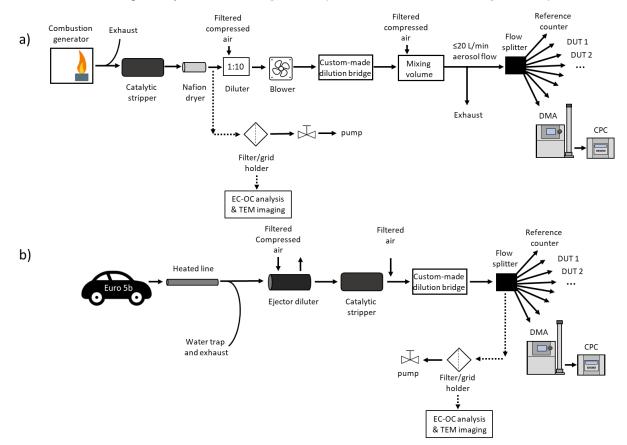
70 During the first measurement campaign at METAS, the following laboratory-based diffusion or premixed flame

- 71 generators were used to produce test aerosols: a prototype bigCAST, a miniCAST 5201 BC (Ess et al., 2021b; Ess
- and Vasilatou, 2019), a miniCAST 6204 C and the miniature inverted soot generator (MISG) (Giechaskiel and
- 73 Melas, 2022; Kazemimanesh et al., 2019; Moallemi et al., 2019; Senaratne et al., 2023). By varying the operation
- points of the CAST generators, polydisperse aerosols with a geometric mean mobility diameter (GMD_{mob}) ranging
- from 50 nm to 100 nm were generated, as summarised in Fig. S1. In the case of the MISG, particles with a GMD_{mob}

- down to 100 nm were produced in a repeatable and stable manner using a mixture of dimethyl ether and propane
- 77 (Senaratne et al., 2023). This is in agreement with another study, where the modal diameter varied between 95 and
- 78 <u>158 nm (Bischof et al., 2020).</u>

79 The counting efficiency profiles (CE) of six DC-based PN-PTI counters, namely the AEM (TEN, the Netherlands), 80 HEPaC (developed by the University of Applied Sciences Northwestern Switzerland and distributed by Naneos 81 GmbH, Switzerland), DiTEST (AVL DiTEST, Austria), CAP3070 (Capelec, France), DX280 (Continental 82 Aftermarket & Services GmbH, Germany) and AIP PDC KG4 (referred to as Knestel hereafter, KNESTEL 83 Technologie & Elektronik GmbH, Germany). The HEPaC, DiTEST, CAP3070 and DX280 had been type-84 approved at METAS according to the Swiss regulations (VAMV, 2018) whereas the Knestel instrument had been 85 type-approved according to the German regulation (AU-Richtlinie, n.d.). The experimental setup at METAS is depicted in Fig. 1a. Soot produced by CAST-burners or the MISG was passed through a catalytic stripper (Catalytic 86 87 Instruments GmbH, Germany), a Nafion dryer (MD-700-12S-1, PERMA PURE, U.S.A.), a VKL 10 diluter (Palas 88 GmbH, Germany) and a custom-made dilution bridge, and was mixed in a 27-ml-volume chamber. The aerosol 89 was split with a custom-made 8-port flow splitter and delivered simultaneously to the devices under test (DUT, in 90 this case PN-PTI instrument) and the reference particle counter (NPET 3795, TSI Inc., USA). The NPET was 91 selected as reference instrument for two reasons; i) it could be used in field measurements as it included a dilution 92 system, a volatile particle remover and a particle counter, ii) during type examination portable PN-PTI instruments 93 are typically used as reference. NPET had been calibrated in a traceable manner according to the ISO 27891

- 94 standard, and showed a CE of 0.77 ± 0.02 , 0.77 ± 0.01 , 0.80 ± 0.01 and 0.79 ± 0.02 at a GMD_{mob} of 50 nm, 70 nm,
- 95 80 nm and 100 nm, respectively and this counting efficiency was taken into account during data analysis.



96

Figure 1: a) Experimental setup for the verification of PN-PTI instruments in the laboratory. Four different combustion
 generators were used (see text for more details). DUT stands for device under test. Dashed arrows designate

- 99 measurements which were performed separately, i.e. not in parallel with PN-PTI verification. b) Experimental setup as 100 used for field measurements at JRC.
- 101 Mobility size distributions were recorded simultaneously by a scanning mobility particle sizer (⁸⁵Kr source 3077A,

102 DMA 3081 and butanol CPC 3776, TSI Inc., USA). To analyse the morphology of the soot particles, particles

- 103 were sampled for 5 s with a flow rate of 1.2 L/min downstream the Nafion dryer, collected on copper-coated TEM
- 104 (transmission electron microscopy) grids placed in a mini particle sampler (MPS, Ecomeasure, France) and
- analysed with a Spirit Transmission EM (Tecnai, FEI Company, USA). Soot particles were also sampled on QR-
- 106 100 Advantec filters (Toyo Roshi Kaisha, Ltd. Japan, preheated at 500 $^{\circ}$ C for > 1 h) for durations of 15 30 min.
- 107 Elemental carbon (EC) to total carbon (TC) mass fractions were measured with an OC/EC Model 5L analyser
- 108 (Sunset Laboratory Inc., NL) by applying an extended EUSAAR-2 protocol (Ess et al., 2021b, 2021a).
- 109 In a second measurement campaign at JRC, the HEPaC, DiTEST, CAP3070 and DX280 counters were challenged
- 110 with real diesel engine exhaust from a Euro 5b vehicle. Fig. 1b depicts the experimental setup at JRC. Soot from
- 111 engine exhaust was passed through a water trap, a heated line (150 °C) to avoid water condensation, an ejector
- 112 dilutor (DI-1000, Dekati, Finland), a catalytic stripper (Catalytic Instruments GmbH, Germany) to remove
- 113 (semi)volatile organic matter, and was diluted to the required concentrations with a custom-made dilution bridge.
- 114 It has been shown that the ejector dilutor does not affect the particle size distribution (Giechaskiel et al., 2009).
- 115 PNC was recorded for several minutes, which allowed identifying long-time trends or drifts of the reported PNC.
- 116 In addition, PNCs were averaged over a period of 1 min, thus the duration was similar to the duration of real PN-
- 117 PTI tests which varies from 15 to 90 s. Mobility size distributions were measured by an SMPS, consisting of an
- ⁸⁵Kr source (3077A, TSI Inc., U.S.A.; purchased in 2021), a DMA 3081 and a CPC 3010 (TSI Inc., USA).
- 119 A Euro 5b vehicle with by-passed DPF was tested as real source of diesel soot. The vehicle generated size
- 120 distributions with a GMD_{mob} of 56.4 nm \pm 0.7 nm. Diesel particles from the Euro 5b vehicle were collected on
- 121 TEM grids and analysed as described above.

122 **3 Results**

123 **3.1 Aerosol properties**

- 124 Particle number concentration measured by diffusion chargers depends on the average number of charges carried
- 125 <u>by each particle (Fierz et al., 2011).</u> Particle size and morphology have been shown to have an effect on the <u>number</u>
- 126 of charges carried by the particles and, thus, on the counting efficiency of diffusion charger based PN-PTI
- 127 instruments (see (Dhaniyala et al., 2011; Vasilatou et al., 2023) and references therein). Soot particles form
- 128 complex structures described by a fractal-like scaling law (Mandelbrot, 1982), and their mobility is influenced by
- 129 their morphology (described by the fractal dimension and fractal pre-factor) and the momentum-transfer regime
- 130 (Filippov et al., 2000; Melas et al., 2014; Sorensen, 2011). To characterise the soot particles produced by the
- 131 different aerosol generators, the following aerosol properties were determined: particle size distribution, EC/TC
- 132 ratio, primary particle size and fractal dimension. EC/TC ratio can also have an effect on the morphology of the
- 133 soot particles. Soot particles formed in premixed flames (i.e. high EC/TC) exhibit a loose agglomerate structure
- 134 where the primary particles are clearly distinguishable from one another, while soot generated in fuel-rich flames
- 135 (high OC/TC) has a more compact structure and the primary particles tend to merge with each other (see Fig. 3 in
- 136 (Ess et al., 2021b)).

137

138 The properties of the soot aerosols are summarised in Table 1. Mobility size distributions are shown in Fig. S1.

139	Table 1: Physical properties of the soot a	erosals produced by the variou	is combustion generators and th	e Fure 5h engine
157	Table 1.1 Hysical properties of the soot a	ici usuis produccu by the variou	is compusition generators and th	ie Euro 50 engine.

Soot	Setpoint	GMD _{mob}	GSD (nm)	EC/TC mass	<i>d</i> _{pp} (nm)**	$ ho_{\rm eff} ({ m g/cm^3})^{***}$	$\underline{D_{\mathrm{f}}}^{\ddagger \ddagger}$
generator		(nm)		fraction (%)*			
MISG	100 nm	103.3	1.76	86.2 ± 10	9.2 ± 2.8	0.91 ± 0.02	$\underline{1.71\pm0.03}$
miniCAST	50 nm	50.7	1.43	57.2 ± 8.9			
6204 C	70 nm	73.4	1.48	27.9 ± 4.6			
	80 nm	80.0	1.54	77.8 ± 9.0			
	100 nm	99.5	1.69	41.9 ± 6.5	21.6±2.5	0.35 ± 0.04	$\underline{1.54\pm0.04}$
miniCAST	50 nm	51.1	1.60	100 ± 18.5			
5201 BC	70 nm	75.3	1.59	94.6 ± 15.6			
	fuel-lean						
	70 nm	74.2	1.69	73.7 ± 11.4			
	fuel-rich						
	80 nm	81.8	1.57	98.1 ± 15.3			
	100 nm	99.8	1.63	97.4 ± 9.6	$15.8\pm3.5^\dagger$	~ 0.4 [†]	$\underline{1.58\pm0.01}$
	fuel-lean						
	100 nm	101.9	1.58	65.7 ± 10.0	Primary	$1.04\pm0.16^{\dagger}$	$\underline{1.71 \pm 0.01}$
	fuel-rich				particles are		
					partly merged [†]		
bigCAST	50 nm	52.5	1.57	50.9 ± 11.7			
	70 nm	71.6	1.54	62.2 ± 13.3			
	80 nm	81.5	1.53	81.2 ± 8.8			
	100 nm	98.9	1.60	100.0 ± 9.0	24.5 ± 1.8	0.66 ± 0.04	$\underline{1.66\pm0.02}$
Vehicle		56.4	2.12 ± 0.00	83.5 ± 20.5	19.7 ± 4.4		$\underline{1.67\pm0.03}$
Euro 5b							

140

* Uncertainties of the EC/TC mass fraction (downstream of the CS) are estimated to be in the range of 10-15 %.

141 Uncertainties due to the split point could not be quantified and were not taken into account.

** Expanded uncertainty (k = 2,95 % confidence interval) determined as the twofold standard deviation of d_{pp} , of 142

143 at least 20 primary particles of various mature soot particles divided by the square route of the number of

144 measurements.

145 *** Expanded uncertainty (k = 2,95 % confidence interval) determined as the twofold standard deviation of three 146 measurements.

147 † Taken from (Ess et al., 2021b).

 $\frac{1}{2}$ Expanded uncertainty (k = 2, 95 % confidence interval) determined as the twofold standard deviation of at least 148

- 149 10 measurements.
- 150

The fractal dimension D_{f} of soot particles with a nominal GMD_{mob} of 100 nm was derived via image analysis of 151

152 high-quality TEM-images using the FracLac feature of ImageJ 1.53e (ImageJ, National institutes of Health, USA). 153 In a first step, the greyscale TEM-images were converted into binary images utilizing the auto-convert function of

154 FracLac. In a second step, the $D_{\rm f}$ values were determined via the so-called box counting, averaging 12 rotations of

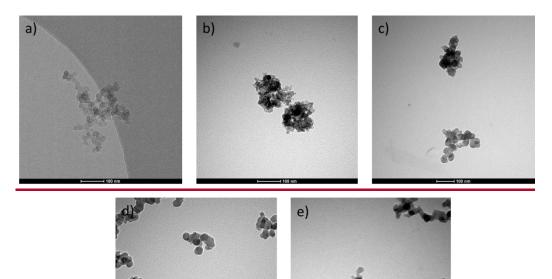
155 <u>each image. The D_{f} values summarised in Table 1 represent the average values obtained from at least 10 particles</u>

156 for each type of soot. These values agree well with those reported in previous studies for bare (i.e. freshly emitted)

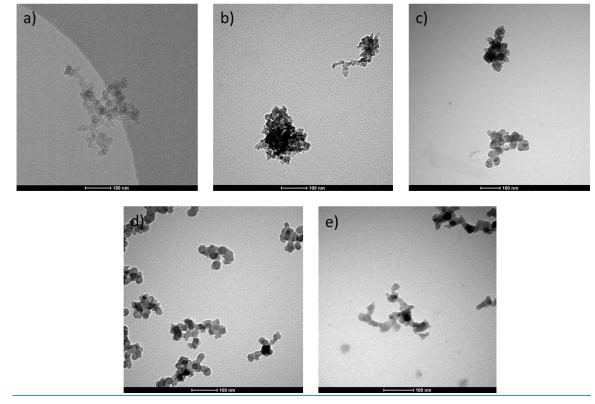
157 <u>soot particles (Pang et al., 2022; Wang et al., 2017).</u>

158 The effective density was determined for the 100 nm setpoints using an Aerodynamic Aerosol Classifier (AAC, 159 Cambustion, UK) and a DMA (TSI Inc., USA) as described in (Tavakoli and Olfert, 2014). The lowest effective 160 density $(0.35 \pm 0.02 \text{ g/cm}^3)$ was found for particles generated by the miniCAST 6204 C. Considering that these 161 particles contain a high amount of OC, this value might seem at first glance to be low, but can be explained by the 162 highly fractal-like structure of soot (Fig. 2e). In comparison, the miniCAST 5201 BC produced particles with an 163 effective density of 1.04 ± 0.08 g/cm³ when operated under fuel-rich conditions (i.e. high OC mass fraction), which 164 is in line with the more compact structure as shown in (Ess et al., 2021b). Similarly, the MISG generated particles 165 with an effective density of 0.91 ± 0.02 g/cm³. 100 nm particles generated by the bigCAST exhibited an 166 intermediate effective density of 0.66 ± 0.02 g/cm³. The results are in line with the calculated fractal dimension of soot particles, which increases from 1.54 for soot generated by the miniCAST 6204 C to 1.71 for soot generated 167

168 by the MISG.



169



171Figure 2: TEM images of polydisperse soot particles generated by a) the miniCAST 5201 BC (GMD_{mob} of ~100 nm, fuel-172lean setpoint); b) the MISG (GMD_{mob} of ~100 nm); c) by the Euro 5b test vehicle (GMD_{mob} of ~55 nm); d) the prototype173bigCAST (GMD_{mob} of ~100 nm); and e) by the miniCAST 6204 C (GMD_{mob} of ~100 nm). Further images are compiled174in Figs. S2-S5 and in (Ess et al., 2021b).

Soot particles generated by the bigCAST with a GMD_{mob} of ~ 100 nm consist of primary particles with a diameter $d_{pp} = 24.5 \text{ nm} \pm 1.8 \text{ nm}$, whereas those from miniCAST 5201 BC (fuel lean setpoint) have an average primary particle size of 12.3 nm ± 3.7 nm at a similar GMD_{mob}. Soot generated by the MISG had a much smaller primary particle size (d_{pp} of 9.2 nm ± 3.8 nm). The TEM images in Figs. 2b and S3 revealed a-that some particles have a more compact soot structure than what reported by (Kazemimanesh et al., 2019) who used ethylene as fuel. This observation is in line with the relatively high particle effective density (0.91 g/cm³) and fractal dimension (1.71) reported above.

182 **3.2** Counting efficiency (CE) profiles of PN-PTI counters

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183 The CE profiles of the PN-PTI instruments under test were determined by dividing the reported number 184 concentration by that measured with a reference condensation particle counter (NPET 3795, TSI Inc., USA). The 185 counting efficiency of the reference counter was taken into account during the data analysis.

- 186 Figure 3 summarises the results obtained with the various laboratory-based combustion generators and the Euro
- 187 5b diesel vehicle. In general, the CE of PN-PTI instruments increased with increasing GMD_{mob}, in line with
- 188 previous studies (Melas et al., 2023; Vasilatou et al., 2023). In the case of CAP3070, CE started to decrease at
- 189 $GMD_{mob} \ge 65$ nm, most probably due to built-in correction factors. It cannot be ruled out that the measurement
- 190 principle of the instrument, based on the so-called escaping current principle, plays also a role (Lehtimäki, 1983).
- 191 In general, for each PN-PTI instrument, the differences in CE when challenged with different soot aerosols of
- similar particle size were <0.25 at 50 nm and increased with size, but remained typically lower than 0.5. Higher
- differences were observed for CAP3070 at around 100 nm, probably related to the internal correction factors. This

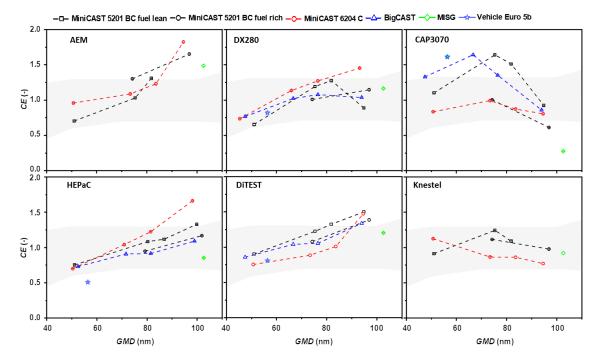
indicates that the exact morphology (e.g. primary particle size, effective density) of the test aerosol had an effecton instrument performance as expected from previous studies (Dhaniyala et al., 2011). The response of each PN-

196 PTI model was, however, individual, making it difficult to draw any general trends. For instance, the CE of the

197 HEPaC was higher when measuring soot particles from the miniCAST 6204 C compared to soot of similar

- 198 GMD_{mob} from the bigCAST. CAP3070 showed the opposite behaviour. At a GMD_{mob} of ~100 nm, DX280
- 199 exhibited a higher CE with soot particles generated by the miniCAST 5201 BC under fuel-rich conditions (i.e.
- 200 lower EC/TC mass fraction) than at fuel-lean conditions (higher EC/TC mass fraction). CAP3070 showed again
- 201 the opposite behaviour. It is also worth mentioning that for the HePAC and DX280 instruments the measured CE
- 202 values scattered more at particle sizes larger than 90 nm. This supports the choice of soot with 50-90 nm mobility
- 203 diameter for the PN-PTI instruments verification linearity tests. The counting efficiency of the different PN-PTI

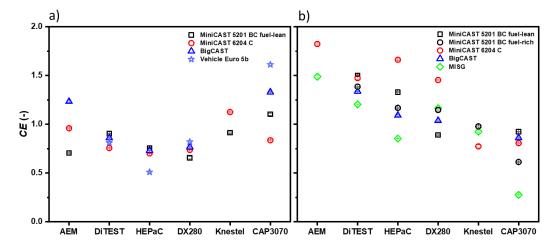
204 counters as a function of time is shown in Figs. S6-S9 for a measurement duration of 2 min.



205

In the case of the DX280 and DiTEST, the CEs reported for the laboratory-generated soot (GMD_{mob} of about 50-55 nm) showed an excellent agreement with the CE measured for diesel soot from a Euro 5b vehicle as shown in Fig. 4a. In all other cases, deviations were observed. These remained typically within 0.25 units in CE but in one case (for CAP3070) reached a factor of 2. Note that for real vehicle exhaust the tolerance (maximum permissible error MPE) according to German regulations is \pm 50% (PTB, 2021). In general, the data indicate that soot produced by miniCAST and bigCAST generators simulate, in most cases, the properties of diesel soot by a Euro 5b vehicle satisfactorily.

Figure 3: Influence of the type of soot generator/vehicle engine (bigCAST, miniCAST 5201 BC, miniCAST 6204 C, MISG and Euro 5b diesel engine) on the counting efficiency (CE) of six different PN-PTI counters: AEM, HEPaC, DiTEST, CAP3070, DX280, and Knestel. The grey-shaded area designates the upper and lower limits in the counting efficiency as defined in the document "Commission Recommendation on particle number measurement for the periodic technical inspection of vehicles equipped with compression ignition engines" (EU, 2023).

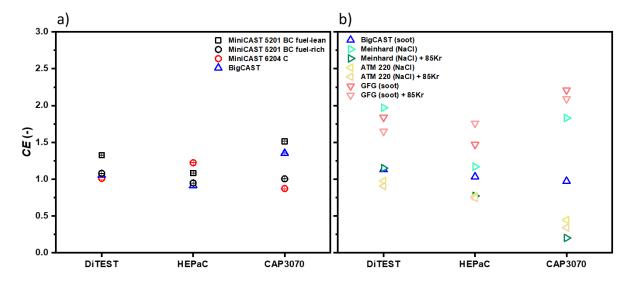




219Figure 4: Influence of the type of soot generator/engine (bigCAST, miniCAST 5201 BC, miniCAST 6204 C, MISG, Euro2205b vehicle) on the counting efficiencies (CE) of six different PN-PTI counters: AEM, HEPaC, DiTEST, CAP3070,221DX280, Knestel (the Knestel and AEM counters were not challenged with the Euro 5b vehicle since the Knestel counter222was sent for service and the performance of the AEM counter deteriorated during the measurement campaign at JRC).223The polydisperse test aerosols had a particle number concentration of ~100'000 cm⁻³ and a GMDmob of a) 50-55 nm and224b) ~ 100 nm.

As shown in Fig 4b, soot generated by the MISG ($GMD_{mob} \sim 100 \text{ nm}$) led to CEs close to 1 for the DX280, DiTEST, Knestel and HEPaC counters, and the CEs lied within the tolerance range defined in Germany and Switzerland (the Netherlands and Belgium only specify a tolerance range for mobility diameters up to 80 nm). The CE limit values were only exceeded in the case of the AEM and CAP3070 counters but this was most probably due to a deterioration of the performance of the AEM instrument or an underestimated internal correction and an overestimated internal correction factor in the case of CAP3070. Although the size of the soot generated by the MISG ($GMD_{mob} \ge 90 \text{ nm}$) tends to be larger than real soot from diesel engines (Kazemimanesh et al., 2019;

- Moallemi et al., 2019; Senaratne et al., 2023), it's ease of operation combined with the affordable price make it an
- attractive choice for PN-PTI verification in the laboratory.



234 235

Figure 5: a) Influence of different soot aerosols with a GMD_{mob} of ~80 nm on the counting efficiencies (CE) of three different PN-PTI counters. b) Influence of different test aerosols (soot, NaCl and carbonaceous particles from a sparkdischarge generator) on the counting efficiencies (CE) of the same PN-PTI counters. The test aerosols had a GMD_{mob}

239 of ~80 nm. The data points are taken from (Vasilatou et al., 2023).

- 240 The variation in the counting efficiency of the PN-PTI instruments when tested with soot particles from different
- combustion generators (Fig. 5a) is much smaller than that observed with test aerosols such as NaCl or particles
- from a spark-discharge generator with a similar GMD_{mob} (Fig. 5b) (Vasilatou et al., 2023). For instance,

243 carbonaceous particles from a GFG spark-discharge generator (Palas GmbH, Germany) led to a CE of ≥ 2 in the

- case of CAP3070 and 1.7-1.8 in the case of DiTEST. On the contrary, CE remained typically in the range 0.7-1.3
- 245 when soot was used as test aerosol, irrespective of the type of combustion generator (Fig. 5a). Further studies with
- 246 more diesel test vehicles would be necessary to elucidate which type of laboratory-generated soot is the best proxy
- for diesel soot, keeping in mind that the properties of real diesel soot can also differ considerably, depending on
- the engine design, driving cycle and fuel properties (Hays et al., 2017; Wihersaari et al., 2020).

249 4 Recommendations

- 250 Based on the results of this study, the following recommendations can be made:
- Initial and follow-up verification of DC-based PN-PTI counters should ideally be performed with soot as test
 aerosol. If possible, the same type of combustion generator should be used for the determination of CE during
 type-examination and verification.
- Low-cost soot generators can be a stable source of combustion particles and can be employed for PN-PTI
 verification using the appropriate setup correction factors. However, the GMD they produce should be in the
 range 70±20 nm in order to comply with the current linearity verification requirements in Europe.
- Laboratory procedures for PN-PTI type-examination and verification should be further harmonised in Europe
 to avoid inconsistencies in the enforcement of PTI legislation. International round robin tests should be
 performed to examine whether a) the various PN-PTI instruments type-examined and verified in different
 European countries according to national regulations exhibit a similar performance and b) whether PN-PTI
 instruments verified in the same country but with different test aerosols identify defect DPFs in a consistent
 manner.
- As highlighted in our previous study (Vasilatou et al., 2023), "setup correction factors" should be determined 263 264 whenever verification is performed with particles other than soot to account for the effects of the test aerosol on 265 the instrument's counting efficiency. These "setup correction factors" depend on both the aerosol physicochemical 266 properties and the instrument's design, and need to be determined at the NMI level at regular intervals as drifts in the performance of the aerosol generator may occur. If "setup correction factors" are not applied or are inaccurate, 267 the reliability of PTI will be compromised. The use of "setup correction factors" is more critical when nebulisers 268 269 or spark-discharge generators are used, but special care should also be given to different flame soot generators. 270 This calls for a closer collaboration between NMIs, state authorities, instrument manufacturers and verification 271 centres to ensure fair implementation of regulations in Europe. Further harmonisation of the different PN-PTI 272 type-examination procedures in Europe, e.g. in terms of the combustion generator, would be a valuable first step 273 in order to determine meaningful correction factors for other test aerosols.

274 5 Conclusions

The type of soot aerosol affected the response of six different DC-based PN-PTI counters tested in this study. Size and physicochemical properties of the test aerosol had effects on the CE of all counters. In most cases, the different

- 277 laboratory-generated soot aerosols resulted in deviations of 0.25 units in the counting efficiency of individual
- counters compared to Euro 5b diesel soot at similar mobility diameters (~50-60 nm). It is not entirely clear which
- type of laboratory-generated soot is the best proxy for real soot emitted by diesel vehicles as the response of the
- 280 PN-PTI instruments to the different test aerosols was not uniform. It must also be kept in mind that the properties
- of diesel soot may vary depending on the engine specification and operation. Nevertheless, this study confirms
- that soot aerosols, irrespective of the generator model, are more suitable as test aerosols than NaCl, oil or particles
- from spark discharge generators. In view of these results, recommendations were made with regard to PN-PTI
- type-examination and verification.

285 Author contribution

- All authors designed the experiments. TH, DR and AM carried out the measurement campaigns. TH analysed the
- 287 data with support from DR. KV prepared the manuscript with contributions from all co-authors.

288 **Competing interests**

289 The authors declare no competing interests.

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