

Effects of the test aerosol on the performance of periodic technical inspection particle counters

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ABSTRACT

A new scheme for periodical technical inspection (PTI) of diesel-engine exhaust based on the measurement of particle number (PN) concentration was recently introduced in several European countries. PN concentration is measured directly at the tailpipe using portable particle counters known as PN-PTI. The technical specifications of these instruments are defined at a national level, and the type-examination and verification procedures differ considerably among European countries. In this study, we examined how the choice of test aerosol can affect the counting efficiency profile of the PN-PTI sensors, the majority of which are based on diffusion charging (DC). Carbonaceous aerosols were produced by combustion and spark discharge generators, sodium chloride (NaCl) particles were generated from aqueous solutions using several commercially available nebulizers and silver particles were produced by a silver particle generator. In most cases, the response of DC-based sensors depended strongly on the morphology of the test particles. Comparison with a Euro 3 diesel engine without a diesel particle filter (DPF) indicated that the combustion aerosols generated by combustion generators were the best proxy for soot emitted by diesel vehicle engines. On the contrary, the use of NaCl and spark discharge particles as test aerosols led in some cases to an over- or underestimation of the PN concentration by up to about a factor of two. Moreover, our results showed that the response of each type of DC-based sensors is individual and, as a result, it is impossible to determine a generally applicable factor to correct for the impact of the test aerosol properties. We recommend that the verification of PN-PTI instruments be preferably performed with soot particles produced by a combustion generator (identical or similar to the one used during the type-examination) in order to avoid corrections for the impact of the material on the response of the instruments.

1. Introduction

Soot particles, formed by the incomplete combustion of carbon-containing fuels or biomass, have received increased attention from the scientific community, society, and policy makers due to their adverse health effects and contribution to global climate change

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(Kheirbek, Haney, Douglas, Ito, & Matte, 2016; USEPA, 2019, 2021; WHO, 2021).

In most urban regions worldwide, on-road vehicles contribute significantly to air pollution. This calls for a more effective control of traffic-related particle emissions (Gong & Zhou, 2018; Kheirbek et al., 2016; Rahman, Mazaheri, Clifford, & Morawska, 2017). Equipping diesel engines with diesel particle filters (DPFs) was a major advance in emission control; however, in some cases DPFs cannot ensure efficient particle removal over the whole service life of a vehicle. Studies have shown that DPFs can be tampered or might simply be prone to defects (Giechaskiel et al., 2022)(H. Burtscher, Lutz, & Mayer, 2019). In these cases, particle emissions from vehicles equipped with defective DPFs can easily exceed the type-approval limit (Giechaskiel, Lähde, Suarez-Bertoa, Valverde, & Clairotte, 2020; Melas, Selleri, Suarez-Bertoa, & Giechaskiel, 2022; Yamada, 2019; Yang et al.,). A small fraction ($\approx 10\%$) of vehicles can be responsible for up to 80–90% of the total fleet emissions (H. Burtscher et al., 2019; Kurniawan & Schmidt-Ott, 2006). Existing methods for vehicle emission control, such as opacity (smoke) meters and on-board diagnostic (OBD) systems, fail to detect high particulate emissions caused by defective or removed DPFs (Bouveroux et al., 2019; Melas et al., 2022).

To reduce particulate emissions from diesel engines equipped with a DPF, a new procedure for the periodic technical inspection (PTI) of vehicles based on particle number (PN) concentration measurement at low or high idling has recently been defined in Switzerland, Germany, Belgium and the Netherlands (ASTRA, 2022; “Regeling voertuigen 2019/328/NL,” 2019; “Richtlinie für die Durchführung der Untersuchung der Abgase von Kraftfahrzeugen nach Nummer 4.8.2 der Anlage VIIIa StVZO und für die Durchführung von Abgasuntersuchungen an Kraftfahrzeugen nach § 47a StVZO ((AU-Richtlinie Stand 2021)),” n.d.; “SR 741.437 - Verordnung des UVEK vom 21. August 2002 über Wartung und Nachkontrolle von Motorwagen betreffend Abgas- und Rauchemissionen,” n.d.). PN-PTI instruments are based on diffusion chargers (DCs) or condensation particle counters (CPCs) which measure PN concentration directly at the exhaust or tailpipe and are highly sensitive to (soot) particles even at sizes smaller than 100 nm. The type-examination procedures for these PN-PTI devices require laboratory-generated soot particles with realistic properties, i.e. particles with a mobility diameter ≤ 200 nm. The particles should be thermally stable at the conditions of the PN-PTI devices, i.e. at heating up to 350 °C in some cases. Although not specifically required by legislation, a high black carbon content (ideally $>80\%$) is recommended in order to simulate the properties of soot generated by motor vehicles (Matti M. Maricq, 2014); a requirement also for the aviation particle number systems (Durdina et al., 2016). National regulations for the yearly verification of the PN-PTI instruments are on the contrary open to the use of other test aerosols, such as sodium chloride (NaCl) particles and carbonaceous particles produced by spark discharge generators. In this study, we examine the effects of different test aerosols on the counting efficiency of.

- i) four newly developed DC-based PN-PTI counters, namely the DiTEST*¹ (AVL DiTEST, Austria), CAP3070 (Capelec, France), AEM (TEN, the Netherlands) and HEPaC (developed by the University of Applied Sciences Northwestern Switzerland and distributed by Naneos, Switzerland),
- ii) the CPC-based Model 3795 NPET PN-PTI sensor (TSI Inc., U.S.A.)
- iii) the MPEC+ (Dekati, Finland), which is a DC-based PN-PEMS (Portable Emissions Measurement System), and
- iv) the APC particle counter (AVL, Austria), which has been designed to measure particle number concentrations of diluted exhaust gas in a way that complies with the PMP (Particle Measurement Programme) and UNECE-R83 requirements.

Further, we discuss the results in the context of the different national legislations and make recommendations for the correct implementation of the various calibration and verification procedures in the laboratory.

2. National regulations

In Switzerland, the technical requirements for PN-PTI instruments are described in Annex 4 of the *Ordinance of the FDJP on Exhaust Gas Analysers* (SR 941.242; in German also known as VAMV) (VAMV, 2018). According to this ordinance, PN-PTI instruments are type-approved in the laboratory with polydisperse soot aerosols of geometric mean mobility diameter GMD_{mob} between 23 nm and 200 nm. The Netherlands and Belgium, two countries that have harmonised their national legislation (“Belgisch Staatsblad 24255,” 2022; “Belgisch Staatsblad 24273,” 2022; “Belgisch Staatsblad 35106,” 2022; “Regeling voertuigen 2019/328/NL,” 2019), require that the counting efficiency of the PTI instruments be determined with monodisperse soot aerosols of mobility diameter d_{mob} between 23 nm and 80 nm (all other tests are performed with polydisperse aerosols). Germany has aligned its national requirements with those for Portable Emission Measurement Systems (PEMS) introduced in 2017 by the European Commission for Real Driving Emissions (Giechaskiel, Bonnel, Perujo, & Dilara, 2019). PN-PTI instruments must fulfil the requirements described in the following two documents: *German PTI directive* (“AU-Richtlinie”) (“Richtlinie für die Durchführung der Untersuchung der Abgase von Kraftfahrzeugen nach Nummer 4.8.2 der Anlage VIIIa StVZO und für die Durchführung von Abgasuntersuchungen an Kraftfahrzeugen nach § 47a StVZO ((AU-Richtlinie Stand 2021)),” n.d.) and the *PN counter specification PTB-A 12.16* (PTB, 2021). The latter requires that the counting efficiency be determined with monodisperse soot aerosols of d_{mob} between 23 nm and 200 nm. Additionally, the instruments must fulfill a $\pm 25\%$ error limit when tested with polydisperse aerosols over the entire size range. National legislations differ in several other aspects, including the minimum measurement range of the PN-PTI instruments and the testing procedures, as explained in (Melas, Selleri, Suarez-Bertoa, & Giechaskiel, 2021; Vasilatou et al., 2022). Table 1 summarises the different requirements regarding particle size and composition of the test aerosol, minimum measurement range and counting efficiency profile of the PN-PTI instruments. In all

¹ This device was developed to meet the specifications defined in Switzerland (VAMV, 2018) and the Netherlands (“Regeling voertuigen 2019/328/NL,” 2019). A modified instrument version that meets the German specifications has also been developed but was not tested in this study.

mentioned countries, the counting efficiency profile of the PN-PTI instruments is determined with combustion particles generated by a CAST burner. These particles are fractal-like, have been found to simulate satisfactorily the physicochemical properties of soot from diesel engines (Ess et al., 2021; Ess & Vasilatou, 2019) and are commonly used to test/calibrate a variety of aerosol measuring instruments (Durdina et al., 2016; Giechaskiel et al., 2012; Kalbermatter et al., 2022; Noble, Desgroux, & Focsa, 2017) (Giechaskiel, Cresnoverh, Jörgl, & Bergmann, 2010). On the contrary, NaCl particles have a crystalline structure when in dry state (Bladt, Ivleva, & Niessner, 2014; Tumolva et al., 2010; Weis & Ewing, 1999; Wise et al., 2007). Carbonaceous particles produced by spark discharge generators are fractal-like agglomerates of very small primary particles (typically ≤ 10 nm) with a low fractal dimension, resulting in a lower effective density than combustion particles by diesel engines (Gysel et al., 2012; Heinz Burtscher, 2020; Müller et al., 2005). Even of higher importance is to understand the effect of the material and particle types on the response of the instruments used for PTI. For example, CPCs with a 50% cut-off at 23 nm have different detection efficiencies at their lower detection limit for different materials (e.g. (Mamakos, Giechaskiel, & Drossinos, 2013)), and in some cases (e.g. NaCl, tetracontane) even for their plateau region (Giechaskiel et al., 2009; Kiwull, Wolf, & Niessner, 2015). DCs can also be affected by the fractality of the particles (Shin et al., 2010). The studies that have examined such effects on PTI counters are limited (Schriefl, Nishida, Knoll, Boies, & Bergmann, 2020), especially when compared with instruments used for the type-approval of the vehicles (Melas et al., 2021)

After the type-examination of a PN-PTI instrument, for each new unit produced by the manufacturer an initial verification applies. Metrological controls apply also to in-use PN-PTI instruments, which are yearly checked through a procedure called subsequent verification. Although in Germany, the Netherlands and Belgium the counting efficiency of PN-PTI instruments is determined with monodisperse aerosols produced by a CAST burner, the (yearly) verification may be carried out with various polydisperse test aerosols at the instrument manufacturers' facilities or other authorized laboratories as summarised in Table 2.

3. Materials and methods

Two measurement campaigns were performed at JRC and METAS, respectively. The JRC campaign involved mostly instruments type-examined in the Netherlands while the METAS campaign focussed exclusively on instruments which were type-examined according to the Swiss regulation. The experimental setup at JRC is depicted schematically in Fig. 1a (testing with polydisperse aerosols) and 1b (testing with monodisperse aerosols). Polydisperse aerosols were generated by the APG particle generator (AVL, Austria) which is a modified miniCAST (model 6.203 C, Jing Ltd., Zollikofen, Switzerland) combined with thermal pre-conditioning and dilution unit (thermal treatment at 350 °C), a DNP 3000 spark discharge generator (Palas, Germany), a Silver Particle Generator (SPG) (Catalytic Instruments, Germany) (Hammer et al., 2022), the salt ATM 228 (Topas, Germany) and salt FCS 249 nebulizers (Topas, Germany), and a Euro 3 diesel vehicle without a Diesel Particle Filter (DPF). The thermal pre-treatment of APG generated particles was used to remove any volatile particles that may be generated at lower GMDs due to the generation method that requires premixing of the propane fuel with nitrogen (Ess & Vasilatou, 2019; Mamakos, Khalek, Giannelli, & Spears, 2013). Testing only with solid particles was considered necessary in order to avoid any size change due to volatile material removal from the PN-PTI counters and also due to the absence of volatile removal devices upstream the reference instrumentation. The salt ATM 228 nebulizer was part of the integrated PN-PTI calibrator of Capelec (France). When needed, the aerosols were diluted with an ejector DI-1000 dilutor (Dekati, Finland) and delivered to the measurement devices. The geometric mean mobility diameter of the aerosol size distribution (GMD_{mob}) was measured with a Scanning Mobility Particle Sizer (SMPS) consisting of Model 3080 electrostatic classifier with a long DMA 3081 (differential mobility analyser) coupled to a Model 3010 butanol 10 nm CPC (TSI Inc., U.S.A.). The reference standard for PN concentration consisted of a dilution unit (Model DDS 560, TOPAS, Germany) and a Model 3752 butanol CPC with a 50% cut-off at 4 nm (TSI Inc., USA). This setup was used to test the following devices under test (DUT):

Table 1

Measurement range of PN-PTI instruments, counting efficiency profile with error limits and test aerosol type for PN-PTI type-examination according to different national legislations.

Switzerland (VAMV, 2018)		The Netherlands & Belgium ("Belgisch Staatsblad 24255," 2022; "Belgisch Staatsblad 24273," 2022; "Belgisch Staatsblad 35106," 2022; "Regeling voertuigen 2019/328/NL," 2019)		Germany ("Richtlinie für die Durchführung der Untersuchung der Abgase von Kraftfahrzeugen nach Nummer 4.8.2 der Anlage VIIIa StVZO und für die Durchführung von Abgasuntersuchungen an Kraftfahrzeugen nach § 47a StVZO ((AU-Richtlinie Stand 2021)), n.d.)	
Minimum measurement range (cm ⁻³)		Minimum measurement range (cm ⁻³)		Minimum measurement range (cm ⁻³)	
50'000 – 5'000'000		5'000 – 5'000'000		5'000 – 500'000	
Particle size GMD_{mob} (nm)	CE (%)	Particle size d_{mob} (nm)	CE (%)	Particle size d_{mob} (nm)	CE (%)
23	<50	23	20 – 60	23	20 – 60
41	>40	50	60 – 130	30	30 – 120
80	70–130	80	70–130	50	60–130
200	<300			70	70–130
				100	70–130
				200	50–200
Typical test aerosol for determining counting efficiency profile bigCAST (polydisperse soot)		miniCAST (monodisperse soot)		miniCAST (monodisperse soot)	

Table 2

Measurement range of PN-PTI instruments, counting efficiency profile with error limits and test aerosol type for PN-PTI (yearly) verification according to different national legislations.

Switzerland		The Netherlands & Belgium		Germany	
Particle size GMD _{mob} (nm)	Error limits CE (%)	Particle size d _{mob} (nm)	Error limits CE (%)	Particle size d _{mob} (nm)	Error limits CE (%)
80	±30	80	±25	70 ± 20	±75
Typical test aerosol for PN-PTI verification soot aerosols; polydisperse Verification institute/center Federal Institute of Metrology (METAS)		NaCl or soot particles; polydisperse Authorized laboratories (private sector)		Spark discharge, soot or NaCl aerosols; polydisperse Authorized laboratories (private sector)	

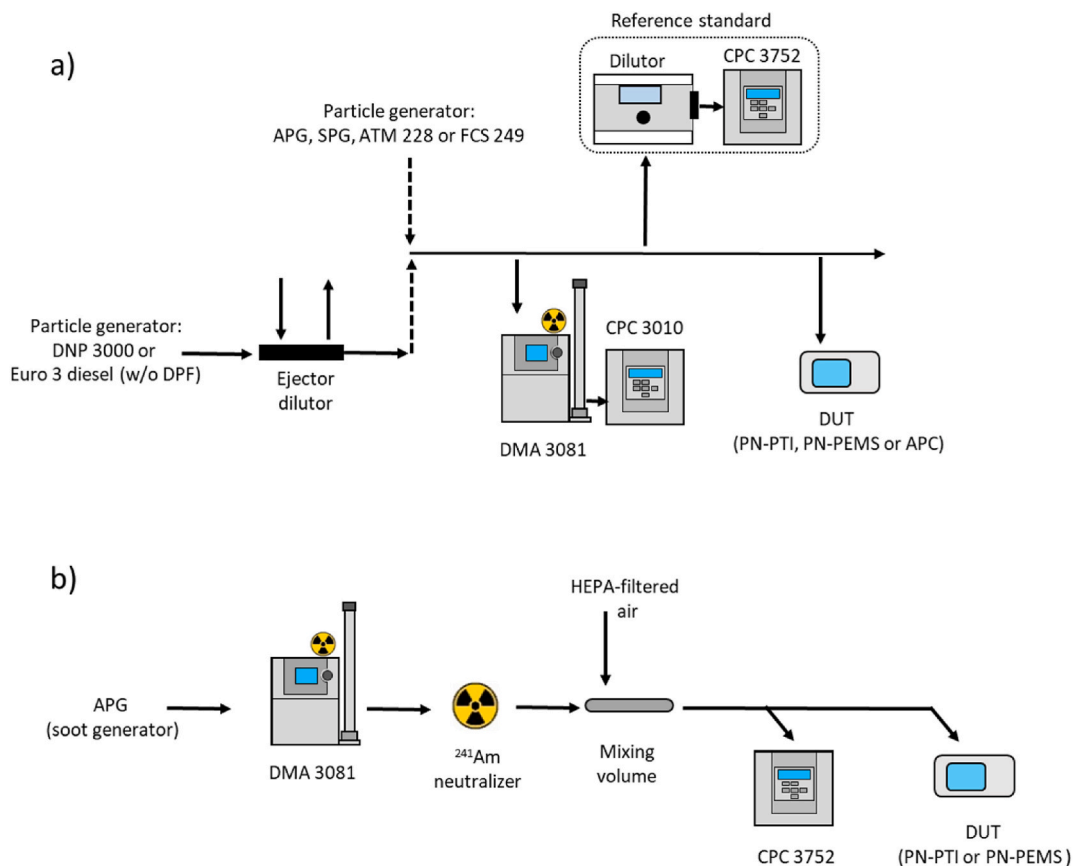


Fig. 1. Schematic illustration of the experimental setup at JRC for measurements with a) polydisperse aerosols and b) monodisperse (i.e. size-selected) aerosols. The dashed lines indicate that the test aerosols were not generated simultaneously. CPC, DMA and SPG and DUT stand for condensation particle counter, differential mobility analyser, silver particle generator and device under test, respectively.

- a) MPEC+ (DC-based PN-PEMS),
- b) DiTEST, CAP3070, AEM (all DC-based PN-PTI sensors),
- c) 3795 NPET (CPC-based PN-PTI sensor).

The same setup was also used to test the APC which incorporated a CPC with a 50% cut-off at 23 nm (hereafter referred to as APC23); a PMP laboratory system for type-approval of vehicles. The latter was traceably calibrated within one year from the start of the testing campaign according to the 2017/1151 requirements. CPC 3752 had a calibration certificate issued within 12 months from the testing campaign. Prior to the testing campaign, the dilution factor of the reference standard dilutor was determined by measuring the soot particle concentrations upstream and downstream the dilutor. Moreover, an inter-comparison of the reference standard (dilutor and CPC 3752) and the APC23 was performed. A detailed characterization of the APC23 can be found elsewhere (Giechaskiel & Melas, 2022). For experiments with monodisperse aerosols, particles generated by the APG burner were classified by their electrical mobility using a DMA 3081 (TSI Inc., U.S.A.) and then passed through an Am-241 neutralizer and mixing volume before being

delivered in parallel to the CPC 3572 and DUT.

The experimental setup at METAS is shown in Fig. 2. Test aerosols were produced by four different aerosol generators: a bigCAST burner (prototype CAST), an EPOND MEINHARD® TR + glass nebulizer (type TRP-50-A0.5, Meinhard, U.S.A) equipped with a cyclone, an ATM 220 nebulizer (Topas, Germany) and a GFG 1000 spark discharge generator (Palas, Germany). The two nebulizers were used to generate NaCl particles. The aerosols had a GMD_{mob} of ~75–80 nm while the GSD was ≤ 1.6 in the case of soot (bigCAST), ~1.7 in the case of the GFG and ATM 220 generators and ~2.1 in the case of the Meinhard nebulizer, as measured by SMPS. The concentration of the nebulized NaCl solutions was $\leq 0.9\%$ w/w. In path a), the aerosol was diluted (and thermally treated at 200 °C in the case of soot particles generated by the bigCAST) to a number concentration of approximately $200'000\text{ cm}^{-3}$, and delivered simultaneously to the reference standard and the device under test (i.e. PN-PTI instrument). For specific tests, an ^{85}Kr aerosol neutralizer (3077A, TSI Inc., U.S.A.; purchased in 2021) and an ion trap to capture ions were placed upstream of the measuring instruments to neutralize the aerosol. The aerosol size distribution had a GMD_{mob} of 80 nm.

The reference standard for particle number concentration consisted of a rotating disc dilutor (RDD, custom-made, METAS) and a Model 3752 CPC (TSI Inc., USA). The reference CPC had been previously calibrated against an electrometer according to the ISO 27891 standard (ISO-27891, 2015). The rotating disc dilutor had been calibrated with two CPCs following the procedure for the determination of splitter bias described in the ISO 27891 standard. The sampling flow rate of the CPC was measured with an external mass flow meter (Vögtlin, Switzerland), which was traceable to the national standard for flow. The ambient pressure and temperature measured in the laboratory were used for the conversion of the flow rate to the ambient conditions during measurement (only for the reference CPC). All connecting tubes were made of conductive silicone material (3001788, TSI Inc., U.S.A.) and the length of the tubes from the flow splitter to the measuring instruments was adapted to the respective flow rate to ensure equal diffusion losses. The splitter bias was measured according to the procedure defined in the ISO 27891 standard and was found to be 1 with a relative uncertainty of $<2\%$ (95% confidence interval).

In path b), the aerosols were size-selected by a DMA (3081, TSI Inc., U.S.A.), neutralized by an ^{85}Kr source (3077A, TSI Inc., USA; purchased in 2021), diluted with particle-free air and delivered simultaneously to the reference CPC and the DUT. The reference CPC was the same as in path a). This setup was used to test the following DUT:

- HEPaC, CAP3070, DiTEST (DC-based PN-PTI) and
- Model 3795 NPET counter (CPC-based PN-PTI) as described in Subsections 4.1 and 4.2.

4. Results and discussion

4.1. Effect of test aerosol on counting efficiency of PN-PTI instruments

Four different PN-PTI instruments were calibrated against a reference standard using different (polydisperse) test aerosols as depicted in Fig. 2, path a). The DiTEST, CAP3070 and HEPaC are DC-based instruments whereas the 3795 NPET is a CPC-based instrument. All of these instruments had successfully passed the type-examination according to the Swiss regulation (VAMV, 2018). To distinguish these from similar models type-approved in other countries (even if the hardware of the DC-based sensors is identical, the software and the built-in calibration settings might differ), we will use the index "CH".

The results of the polydisperse test with GMD of 80 nm are summarised in Table S1 and illustrated in Fig. 3. The DC-based PN-PTI instruments exhibited a counting efficiency between 0.9 and 1.10 when tested with combustion particles from a bigCAST generator. This is no surprise, since the specific DC-based devices had been previously calibrated with similar aerosols in order to fulfil the Swiss specifications. The CPC-based NPET had a counting efficiency of 0.8 with bigCAST soot and slightly lower (around 0.7) with carbonaceous GFG particles or slightly higher (0.85) with salt particles. Carbonaceous particles generated by the GFG generator resulted in counting efficiencies of about 1.7–2.2 for the DC-based instruments. Addition of an ^{85}Kr source downstream of the generator had only a small impact on the counting efficiencies (0.1–0.2 units), indicating that the polydisperse aerosol was not significantly charged (net charge). Salt particles (NaCl) had a significant effect on the counting efficiency of the DC-based devices. The results strongly depended on the type of nebulizer² and on the presence/absence of an ^{85}Kr source downstream of the generator, and they were instrument dependent. For example, for CAP3070_{CH}, the counting efficiency was 0.45 with salt particles produced by the ATM 220 nebulizer, but 1.8 in the case of salt particles from the Meinhard nebulizer. The ^{85}Kr had negligible impact on the particles generated by the ATM 220 nebulizer (–0.1 units), but very pronounced on those produced by the Meinhard generator (–1.45 units).

For all DC-based sensors, the results with the Meinhard nebulizer were strongly affected by the ^{85}Kr source, indicating that the initial charge state of particles significantly affects instrument response. In the case of the ATM generator, the presence of an ^{85}Kr source had negligible impact on the counting efficiency of the DC-based sensors (<0.1 units). In general, the counting efficiencies observed with NaCl particles from the Meinhard nebulizer were deviating more from the ideal value of 1 than with the NaCl particles from the ATM nebulizer. The HEPaC_{CH} was affected the least by the salt particles. Experiments performed at four different particle number concentrations in the range $50'000 - 2'000'000\text{ cm}^{-3}$ showed that the linearity of the instruments was in all cases good and that the number concentration did not affect the counting efficiency of the instruments.

² To minimize deviations in instrument response between soot and salt particles, Capelec recommends to nebulize NaCl solutions with a concentration of $\leq 0.9\%$ w/w, a GMD_{mob} of $70\text{ nm} \pm 20\text{ nm}$ and a GSD in the range 1.6–2.1 with a constant flow rate at atmospheric pressure (personal communication, 18 January 2023).

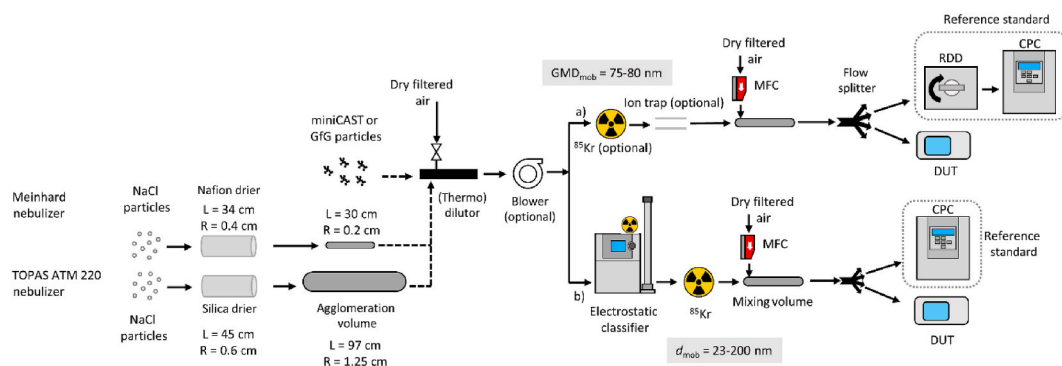


Fig. 2. Schematic illustration of the experimental setup at METAS. This setup was used for experiments with a) polydisperse and b) monodisperse aerosols. RDD, CPC and DUT stand for rotating disc dilutor, condensation particle counter and device under test respectively. The dashed lines designate that either carbonaceous or NaCl aerosols were delivered to the PN-PTI instruments; the test aerosols were never generated simultaneously. L and R denote the length and radius of the driers and agglomeration tubes. CPC, RDD, MFC and DUT stand for condensation particle counter, rotating disk dilutor, mass flow controller and device under test, respectively. GMD_{mob} stands for geometric mean mobility diameter.

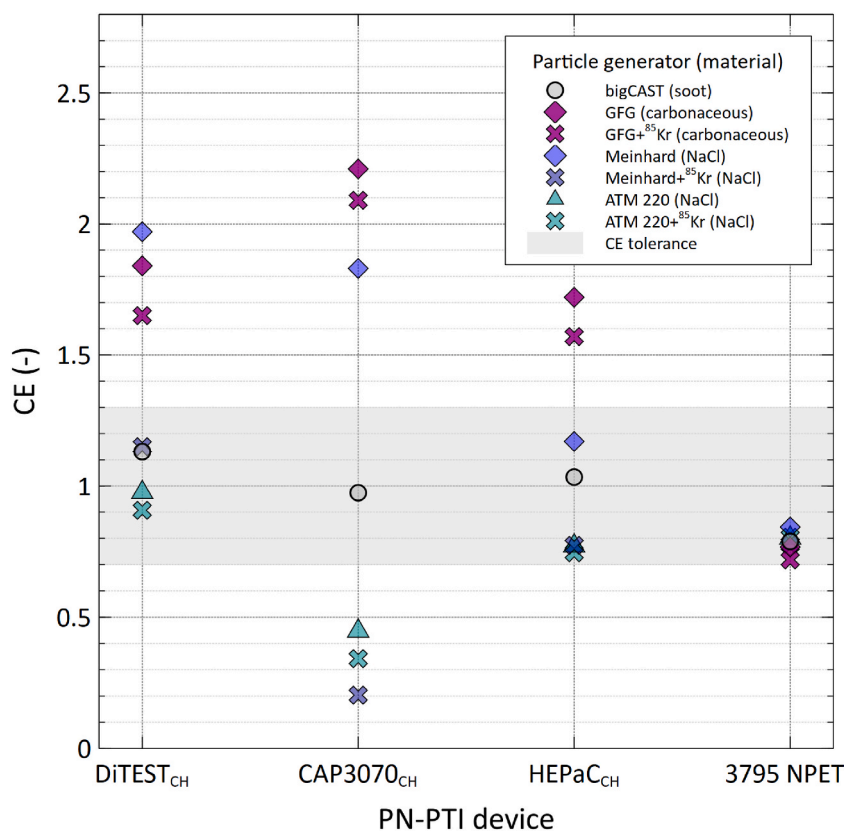


Fig. 3. Effect of the test aerosol on the counting efficiency of four different PN-PTI instruments (the index “CH” indicates that the instruments had already been type approved in Switzerland). The measurements were carried with the experimental setup depicted in Fig. 2 (path a). For all particle generators, the aerosol size distribution had a GMD_{mob} of 75–80 nm and the total number concentration was approximately $200'000 \text{ cm}^{-3}$. The expanded measurement uncertainty (95% confidence level) was typically 4–6% (see Table S1). The error bars are not plotted to improve the readability of the graph. The shaded gray area designates an error limit of $\pm 30\%$ in the counting efficiency, as defined in Switzerland for the PN-PTI verification.

Schrieffl et al. (Schrieffl et al., 2020) found a difference of $\leq 15\%$ between the calibration with soot and salt particles with respect to the total solid particle number concentration in the case of the Partector (Naneos, Switzerland) and a custom-built pulsed-mode modular diffusion charger (PMDC). Our study suggests that each DC-based sensor-type responds differently to the various test aerosols. In order to normalize the responses to “soot” aerosol, different correction factors need to be applied during the yearly verification

depending on the particular characteristics of both the sensor and the aerosol generator. Consistent with previous studies, the counting efficiency of the CPC-based PN-PTI counter used in this study (3795 NPET) did not depend on the material or pre-charge of the test aerosol in the plateau region and no “material” correction would be needed in this case (Tauber, Steiner, & Winkler, 2019).

The TEM images in Fig. 4 show that NaCl particles are compact. The NaCl particles produced by the Meinhard nebulizer are single crystals, while in the case of the ATM 220 nebulizer some of the crystals have agglomerated, probably due to the longer residence time in the agglomeration volume (Fig. 2). The particles produced by spark discharge had the loosest structure and bigCAST particles were somewhere in between. The effect of particle morphology on the charging efficiency has been investigated in several studies, see for example (Oh, Park, & Kim, 2004) and (Jung & Kittelson, 2005). Oh et al. (2004) showed that for particles of the same mobility diameter, particle charge decreased as the sintering temperature in the tube furnace increased and the particles became more compact. Wang et al. (Wang et al., 2010) used the combination of a diffusion charging device with, among others, a CPC to characterize particles in terms of morphology, thus making use of the influence of morphology on the charging efficiency. Useful information on the performance of charge-based instruments can be found in Dhaniyala et al. (Dhaniyala, Fierz, Keskinen, & Marjamäki, 2011). Fierz et al. (Fierz et al., 2016) compared the performance of the diffusion charging device that HEPaC is based on using NaCl and soot particles as test aerosols. The consistent result is that for the same mobility-equivalent diameter the lower the fractal dimension, the higher the charging efficiency. A possible explanation is that the charge-equivalent diameter differs from the mobility-equivalent diameter (Wen et al., 1984). Moreover, as shown in previous studies, particles with a larger dielectric constant can obtain more charges and this effect may increase with lower fractal dimension (Gopalakrishnan, Thajudeen, Ouyang, & Hogan, 2013; Shin, Qi, Wang, Fissan, & Pui, 2009). Consequently, DC-based instruments that measure current of charged particles will have a different response to spheres and agglomerates of the same mobility size. This means that the lowest counting efficiency is expected for NaCl particles and the highest one for particles produced by spark discharge generators. This at least qualitatively explains the results found for particles from the spark discharge generator and the ATM 220. As only little is known on the details of hardware and data evaluation of the devices tested it is impossible to provide any quantitative explanations.

Another important parameter for the response of DC-based instruments is the pre-charge carried by the particles. In the case of the Meinhard nebulizer, the experiments showed a strong difference in the counting efficiency found for NaCl particles with and without aerosol neutralizer. Particles produced by nebulization of salt solutions can be highly charged (the droplets, depending on their size, may acquire charges which remain on the particle when the water evaporates). This charge can lead to significant particle losses and is likely to also affect the reading of instruments depending on the electrical charge the particles carry, see for example Qi et al. (Qi, Asbach, Shin, Fissan, & Pui, 2009). Losses due to high particle charge may also affect CPC-based instruments. Knoll et al. showed that - depending on the operating method of the DC-based instruments - positively pre-charged particles tend to have a larger effect on instrument response than negatively pre-charged particles (Knoll, Schriefl, Nishida, & Bergmann, 2021). This is because negatively

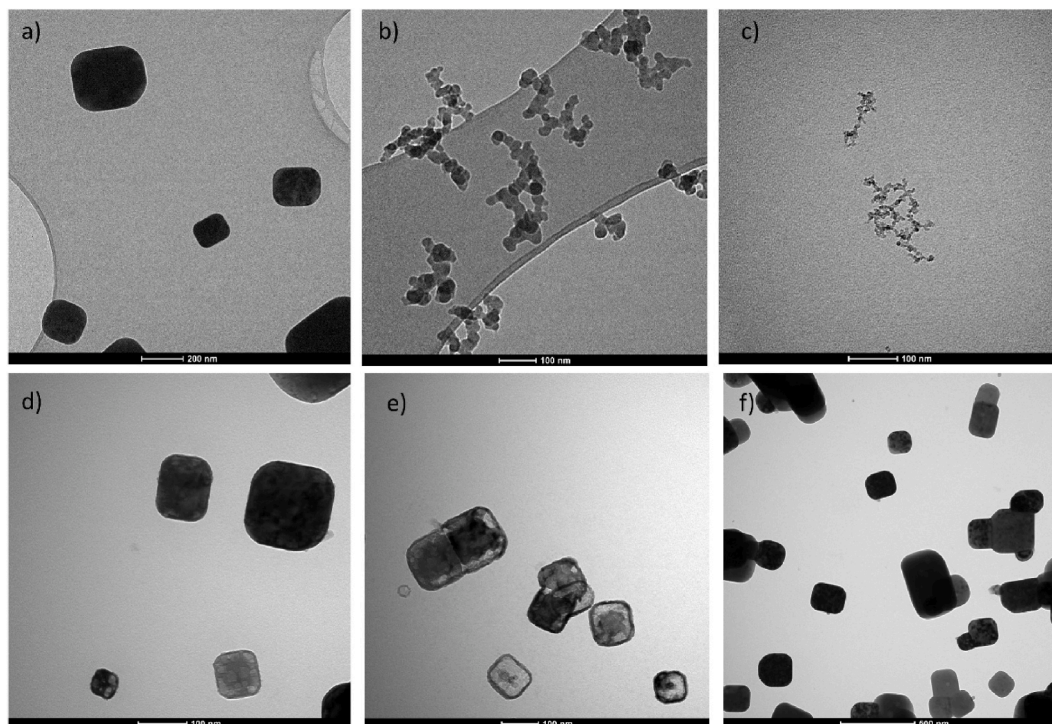


Fig. 4. TEM images of a) NaCl particles generated by the Meinhard nebulizer, b) soot generated by the bigCAST, c) spark discharge particles from the GFG generator and d-f) NaCl particles generated by the ATM 220. The particles were sampled on TEM grids right before the aerosol inlet of the PN-PTI instruments, i.e. after treatment with an aerosol drier or thermodilutor and, in the case of NaCl particles, after the agglomeration volume.

pre-charged particles have very large combination coefficients with the positive ions produced by the unipolar charges due to attractive Coulomb forces. Consequently, the negatively pre-charged particles will become neutral almost immediately (Qi et al., 2009). On the contrary, particles carrying a positive pre-charge will retain these and acquire additional charges when passing through a positive unipolar charger. Further, it is found that the sensitivity to pre-charged particles depends on the charger's $N_{\text{ion}} \cdot t$ product (the product of ion concentration and particle residence time), therefore the response to different aerosol particles with different levels of pre-charging will also depend on the instrument design (Qi et al., 2009). Highly charged particles emitted by Diesel vehicles with Selective Catalytic Reduction (SCR) have been reported in the literature when SCR temperature overcomes 300 °C (Mamakos, Schwelberger, Fierz, & Giechaskiel, 2019). However, lower temperatures are expected at idling. Again, not enough information on the exact design and operation of the PN-PTI instruments is available to quantitatively explain the observed results, but these seem reasonable for the DiTEST and HEPaC counters. The CAP3070 instrument has probably an internal correction for the mean size of particles, which seems to deviate for highly charged particles.

To gain more insight into the performance characteristics of different PN counters, a second measurement campaign was performed in the GMD_{mob} range 17 nm–110 nm using the setup presented in Fig. 1 a). Fig. 5 presents the counting efficiency of six particle counters (MPEC+, APC23, DiTEST, CAP3070, AEM and 3795 NPET) compared with the reference standard for various GMD_{mob} of the test aerosol. Note that the MPEC+ is a PN-PEMS for on-board measurements and APC23 an instrument used for chassis-dyno type approval vehicle testing. The DiTEST had been type-approved in the Netherlands whereas the CAP3070 was similar but not identical to the model homologated in the Netherlands. The AEM model was a device that was later type-examined in Switzerland. Six different model aerosols were used for the tests as described in Section 3. Soot particles (generated by the APG) covered the GMD_{mob} range 23 nm–110 nm, carbonaceous particles (generated with DNP 3000) the range 29 nm–66 nm, salt particles (generated with FCS249 and ATM 228) the range 55 nm–65 nm, silver particles (generated by SPG) the range 17 nm–29 nm, and finally the diesel exhaust particles had a GMD_{mob} of 65 nm.

The results, shown in Fig. 5 and summarised in Tables S2 and S3, confirm the general trends discussed previously (see Fig. 4): the CAP3070 and DiTEST counters overestimated the number concentration of carbonaceous particles generated by spark discharge (DNP 3000) while other counters, such as the AEM,³ CAP3070 and to a lesser degree the MPEC+ underestimated the number concentration of NaCl particles. In general, silver and soot particles of the same GMD_{mob} resulted in similar CE, with the exception of the CAP3070, which was less sensitive to silver particles. However, silver size distributions only up to 30 nm could be produced and is therefore difficult to extend the method or extrapolate the results to larger particle sizes. Measurements with a Euro 3 diesel engine agreed very well with those of soot aerosols from a combustion generator, confirming that soot from CAST or CAST-based generators, such as the AVL particle generator (APG), simulated well the properties of real diesel engines. In addition to material dependence, DC-based sensors had also a size dependence, which is typical for this measurement technology. With increasing particle size, the CE also increased. For the CAP3070 and AEM instruments we observed a decrease in CE at GMD_{mob} larger than 80 nm. This typically occurs due to a correction of the measured concentration by incorporating a diffusion stage prior to the absolute filter that is connected to the electrometer (Fierz, Houle, Steigmeier, & Burtcher, 2011). This setup can be used to estimate the GMD_{mob} and thus correct the CE for higher GMD_{mob} . This hypothesis could not be confirmed due to limited information on the sensors' hardware. The CPC-based NPET counting efficiency showed neither a material dependence nor a size dependence at the CE plateau region. APC23 exhibited similar counting efficiencies for carbonaceous particles from the DNP 3000 spark discharge generator and soot particles from the APG and the Euro 3 diesel engine. The results are in good agreement with those reported or estimated in the literature (Giechaskiel & Melas, 2022). A strong decrease in counting efficiency was, however, observed for NaCl particles irrespective of the type of nebulizer (FCS 249 and ATM 228). The decreased sensitivity to NaCl particles with a GMD_{mob} of 60 nm is surprising considering that this particle size lies well above the 50% cut-off size of the counters and CE has (almost) reached the plateau level. Nevertheless, a lower counting efficiency with salt as test aerosol compared to other materials has already been reported in the literature, but in a smaller degree (Giechaskiel et al., 2009). More recently, lower counting efficiencies even at large particle sizes were also reported (Barouch Giechaskiel & Melas, 2022; Krasa, Kupper, Schriefel, & Bergmann, 2023). NPET did not show such sensitivity to salt particles. This counter uses an internal dilution with re-circulating air, thus some humidity could still exist in the diluted aerosol. Internal losses in the instruments due to the charged salt particles cannot be ruled out.

This result shows that CPC-based devices with cut-off size of 23 nm have a material dependence, which is more profound near their cut-off size but can also occur at larger sizes. Thus, special attention should also be given also to CPC-based PN-PTI instruments for their material dependence as well as on the reference systems used during metrological verifications.

In summary, Fig. 5 highlights the need of defining a correction factor to be applied to initial/subsequent verifications by the instrument manufacturers with regards to type examination tests by the NMIs. This factor should be a "setup correction factor" and include the effect of material, particle size, and reference system.

4.2. Calibration of PN-PTI instruments with monodisperse soot aerosols versus verification with polydisperse soot

As explained in Section 2 and summarised in Table 1, national regulations set different demands on the CE profile of the PN-PTI instruments. Moreover, type examination in Switzerland is carried out with polydisperse soot aerosols while in the Netherlands/Belgium and Germany CE is determined with monodisperse soot aerosols. Three PN-PTI devices (HEPaC, DiTEST and CAP 3070),

³ Note that AEM offers an additional operation mode optimized for salt particles. In this study, we used exclusively the "official" mode intended for PTI measurements (soot particles) of diesel vehicles.

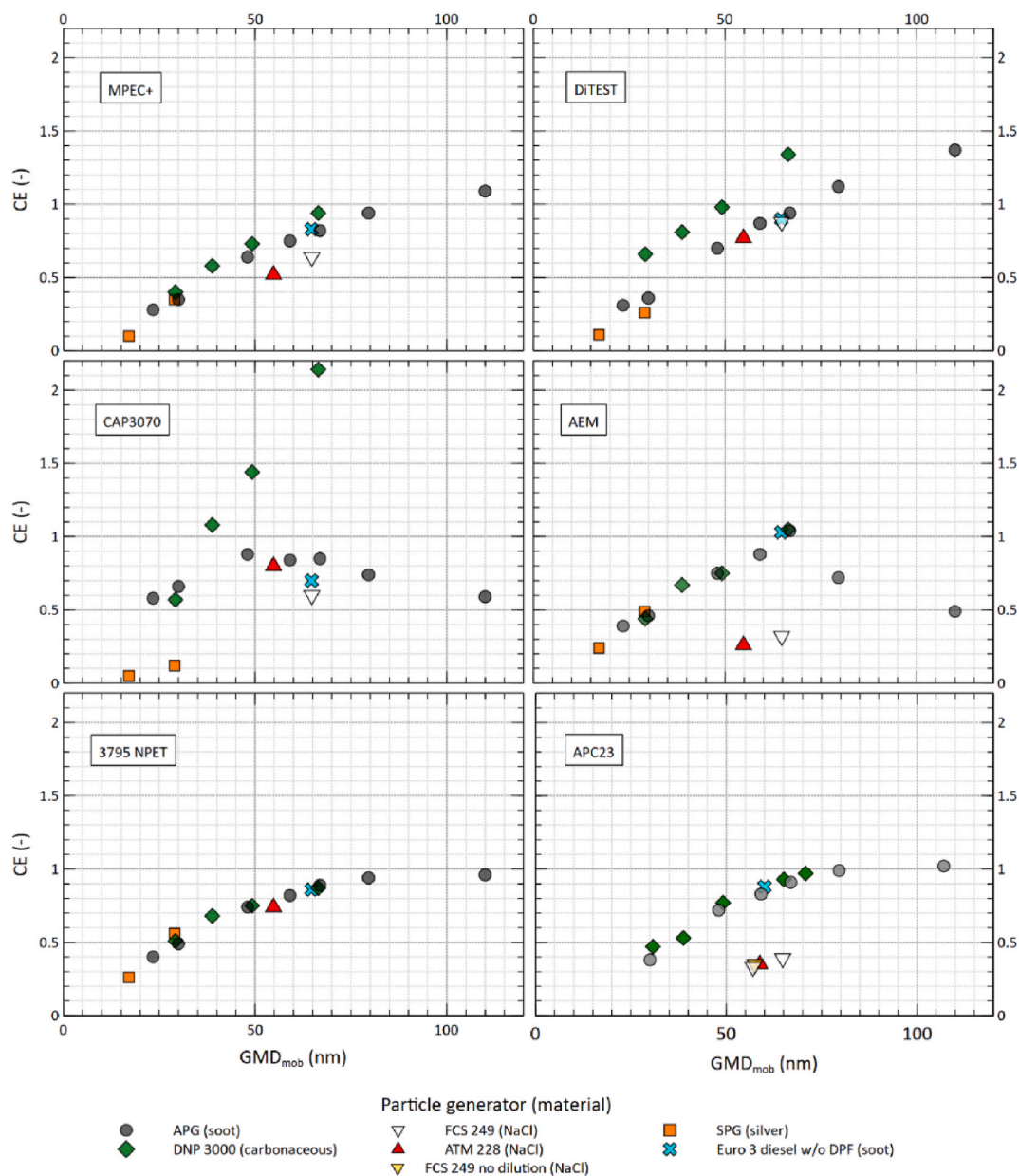


Fig. 5. Counting efficiency (CE) profile of various particle counters depending on the (polydisperse) test aerosol. The measurements were carried out with the experimental setup depicted in Fig. 1 a). The expanded measurement uncertainty (95% confidence level) was typically 8.7%. The error bars are not plotted to improve the readability of the graph. Note that the AEM instrument used in this study was successfully type-examined in Switzerland at a later stage.

which had successfully passed the Swiss type-examination, were tested at METAS with monodisperse and polydisperse soot aerosols according to the German and Swiss technical specifications, respectively. The test aerosols were produced by a bigCAST and the measurements were carried out using the experimental setup illustrated in Fig. 2.

The results are plotted in Fig. 6 and listed in Table S4. The data are presented as solid and open symbols for mono- and polydisperse aerosols, respectively. The measurements revealed that the counting efficiencies of the HEPaC and DiTEST¹ instruments increased faster with increasing soot mobility diameter when challenged with monodisperse aerosols, exceeding the upper CE limit of 2 (200%) at 200 nm set by the German national regulations (PTB, 2021). On the contrary, when using polydisperse soot aerosols, the counting efficiency increased in a more moderate fashion and remained below 2 even at a GMD_{mob} of 200 nm. Furthermore, the counting efficiency of the CAP3070 sensor at 23 nm decreased from 0.34 to 0.10 when switching from polydisperse to monodisperse soot aerosols, thus falling below the lower CE limit of 0.2 set by the German national regulations (PTB, 2021).

To further explore the effect of the aerosol size distribution on the performance of PN-PTI counters, a second measurement

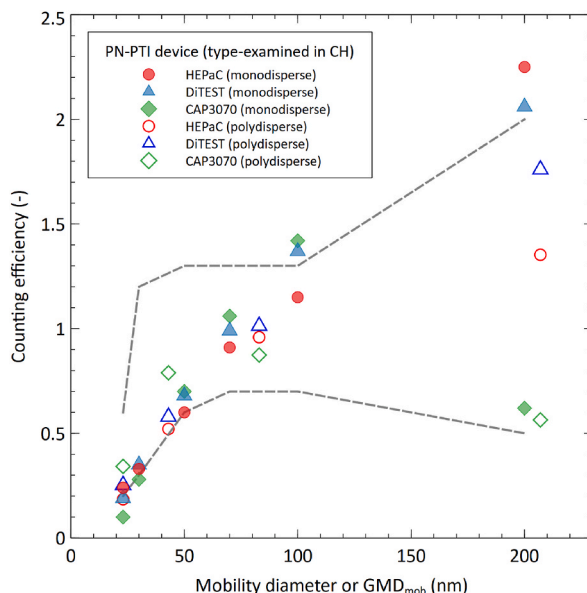


Fig. 6. Counting efficiency profile of the HEPaC, DiTEST and CAP3070 devices with monodisperse (German type-examination) and polydisperse (Swiss type-examination) soot aerosols. The test aerosols were produced by a bigCAST combustion generator and the measurements were carried out with the set up depicted in Fig. 2. The grey dashed lines denote the upper and lower limit in the counting efficiency as defined in the document *PN counter specification PTB-A 12.16* (PTB, 2021). Note that the PN-PTI instruments tested here were the specific models optimized for the Swiss type-examination.

campaign with the DiTEST, MPEC+, AEM, CAP 3070 and NPET counters took place at JRC. The results are depicted graphically in Fig. 7 and listed in Table S5. As previous studies have shown, for systems whose CE depends on the power of the particle diameter (e.g., diffusion chargers) the counting efficiency may differ significantly for monodisperse with respect to polydisperse aerosols at the same particle size, depending on the exponent x ($CE \propto d_{mob}^x$) and the geometric standard deviation of the test aerosol (Fierz et al., 2011; Maricq, 2013). For systems with sigmoid-like CE curves (e.g. CPCs) smaller differences (<0.1 unit) have been found (Giechaskiel & Melas, 2022). In this study, instruments such as the DiTEST, MPEC+ and AEM exhibited a higher CE for monodisperse than for polydisperse aerosols by about 0.1–0.2 units, while for the 3795 NPET the deviation was smaller (typically ≤ 0.1 units). On the contrary, the CAP3070 exhibited a lower CE for monodisperse aerosols by up to 0.4 units for mobility diameters <70 nm but this trend reversed at larger particle sizes.

Note that the results presented in Figs. 6 and 7 and were obtained with soot generated by a bigCAST and an APG, respectively. The APG relies on CAST technology for the combustion of gas fuels, therefore the bigCAST and APG generators contain a similar type of

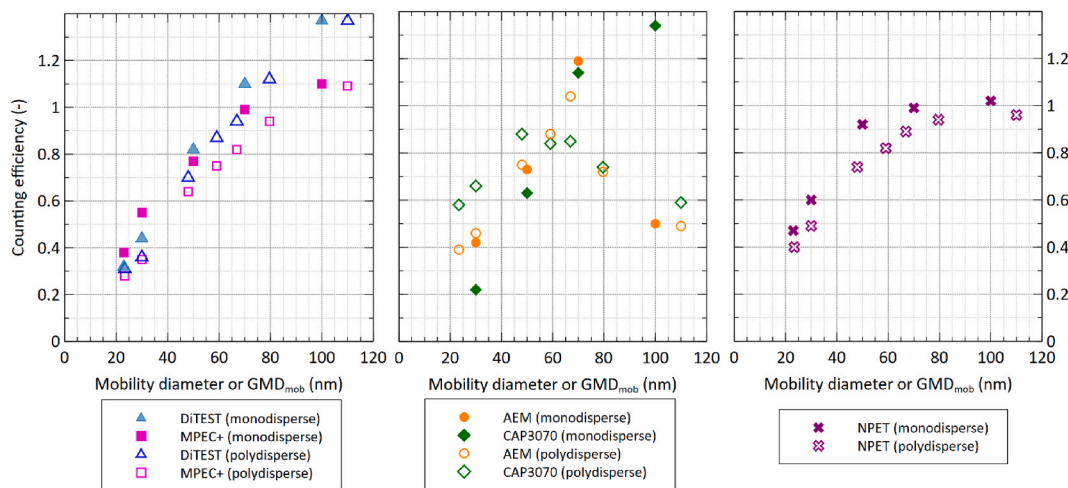


Fig. 7. Counting efficiency profile of the DiTEST, MPEC+, AEM, CAP3070 and 3795 NPET counters with monodisperse and polydisperse soot aerosols. The test aerosols were produced by an APG combustion generator and the measurements were carried out with the set up depicted in Fig. 1 b). The expanded measurement uncertainty (95% confidence level) was 9%.

burner. Preliminary tests indicate that PN-PTI instruments have a similar response to soot generated by the bigCAST and the prototype miniCAST 5201 Type BC (its commercial follow-up model is the miniCAST 5301) (Ess et al., 2021; Ess & Vasilatou, 2019). Further studies are needed, however, to investigate the response of PN-PTI instruments to soot produced by different combustion generators, e. g. by different miniCAST models and the miniature inverted soot generator (Kazemimaneh et al., 2019).

5. Recommendations

In agreement with the results of this study, some recommendations on PN-PTI instruments testing are listed below:

- 1) The results of this study as well as earlier results indicate that the PN-PTI instrument response depends on the type of aerosol used. In all cases the pre-charge carried by the particles may affect the reading of the instrument. The reading of DC-based instruments also depends on particle morphology. Another cross-sensitivity is the chemical composition of the particles, especially for instruments with a 50% cut-off in the counting efficiency at 23 nm. This mainly concerns CPC-based instruments, where the supersaturation needed to initiate particle growth is composition-dependent. For example emery oil, often used for calibration purposes, shows a significantly different behaviour to soot particles (Terres, 2017). As a consequence, it is highly recommended to use particles for calibration purposes which are similar in chemistry and morphology to soot particles emitted by vehicle engines. Special attention should be also given to the graphite generator as in some cases significant differences were observed.
- 2) If verification of PN-PTI instruments is nevertheless performed with test aerosols other than combustion particles, the authors of this study strongly recommend to include in the national (and future European) recommendations the need for calculating an appropriate correction factor for the counting efficiency. The correction applied at initial and subsequent verifications should be done through a “setup correction factor” that includes differences occurring due to different particles material, particle size distribution, and reference system accuracy. Thus, during a PN-PTI instrument initial/subsequent verification, the error calculation should consider the setup correction factor in order to compare with type examination tests. The setup correction factor should be determined at NMI level for the PN-PTI instrument that has passed the type examination tests using the same particle generator that will be used for the initial/subsequent verifications and the reference system that will be used for these verifications. Both DC- and CPC-based PN-PTI instruments should be subject to this test. Preferably, this test should be performed together with the type examination. Any change of the setup (i.e. generator or reference system) should result in a new determination of the “setup correction factor” by the NMI. Further studies are needed to assess the uncertainty of this factor, i.e. drifts over time or when tests are performed at different laboratories.
- 3) Counting efficiency testing is typically performed with monodisperse particles; however, the uncertainty can be very high as the concentrations are close to the detection limits of the instruments. Considering the concentration range of interest for the PN-PTI test ($>10^5 \text{ cm}^{-3}$), polydisperse particles testing should also be considered. Indeed, initial and subsequent linearity verifications are performed with polydisperse particles in all national regulations. According to our study, the difference between monodisperse and polydisperse may vary for different sensors and particle sizes, but, in general, the agreement was satisfactory. There were two exceptions: the CAP3070 sensor at sizes 70–100 nm and HEPaC sensor at 200 nm. For the first instrument, we believe that the differences are due to internal (built-in) corrections for particle size. Our recommendation is that counting efficiency tests may be performed either with monodisperse or polydisperse particles, giving the possibility to the instrument manufacturer to choose the preferred method, if not specified by the relevant (e.g. national) regulation.

6. Conclusions

This study investigated the effects of different test aerosols on the counting efficiency of five PN-PTI sensors (HEPaC, DiTEST, AEM, CAP 3070 and 3795 NPET), a PN-PEMS (MPEC+) and a PMP-compliant particle counter (APC). Carbonaceous aerosols were produced by combustion and spark discharge generators, NaCl particles were nebulized from aqueous solutions and silver particles were produced by a commercial silver particle generator. The response of the DC-based sensors depended strongly on the properties, especially the morphology and the charge, of the test particles. The MPEC + sensor, although DC-based, was less sensitive to the morphology of the test aerosols. Comparison with a Euro 3 diesel engine (without DPF) indicated that (polydisperse) combustion aerosols generated by the bigCAST were the best proxy for soot emitted by diesel vehicle engines. In contrast, the use of NaCl and spark discharge particles as test aerosols led in some cases to a deviation in the reported PN concentration by up to about a factor of two. Moreover, all sensors apart from CAP3070 exhibited a higher counting efficiency for monodisperse than for polydisperse aerosols by up to about 0.2 units. The results indicated that the response of the DC-based sensors to the different test aerosols depends on the particular design and technical characteristics of the sensor. It is therefore impossible to determine a “universal” correction factor to account for artefacts related to the test aerosol properties. We recommend that both type-examination and subsequent verification of PN-PTI instruments be performed with test aerosols that simulate the properties of diesel exhaust, i.e. soot particles produced by a combustion generator.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All measurement results are listed in Supplemental File

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jaerosci.2023.106182>.

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