

Experimental Comparison of Two Portable and Real-Time Size Distribution Analyzers for Nano/Submicron Aerosol Measurements

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ABSTRACT

Two portable, battery powered particle size distribution analyzers, TSI NanoScan scanning mobility particle sizer (TSI NanoScan SMPS 3910, USA) and Kanomax Portable Aerosol Mobility Spectrometer (Kanomax PAMS 3300, Japan), have been recently introduced to the market. Both are compact and allow researchers to rapidly measure and monitor ambient or indoor ultrafine and nanoparticles in real time. In addition, both instruments apply the SMPS measuring scheme, utilizing a corona charger in place of a radioactive neutralizer, and are integrated with a hand-held condensation particle counter (CPC). In this study, the different designs, flow schemes, and operational settings of both instruments have been summarized based on the information released by the manufacturers and the available published literature. The performance characteristics and monitoring capability of these two portable ultrafine to nanoparticle sizers were investigated and compared to a reference TSI 3936 lab-based SMPS under identical conditions. Reasonable agreement was found between the three instruments in terms of their efficiency in sizing and counting polydispersed particles. Of the two portable analyzers, PAMS was able to provide a higher sizing resolution for monodispersed particle measurements than NanoScan, when operated under the High Mode (higher sheath to aerosol flow ratio).

Keywords: Portable aerosol mobility spectrometer; SMPS; Ultrafine particle; Aerosol measurement; Nanoparticle exposure.

INTRODUCTION

The health risks associated with particulate matter (PM) are not only dependent on their composition but also on their size (Cahill and Cahill, 2013). While the PM composition directly corresponds to toxicity, the PM size affects lung capture efficiency (Heyder *et al.*, 1986). Studies have shown that PM_{2.5} (PM with an aerodynamic diameter $\leq 2.5 \mu$ m) is more hazardous than coarser PM due to its longer residence time in the atmosphere and its ability to penetrate deeper into the lungs (World Health Organization, 2006; Li *et al.*, 2013).

More recently, the smaller-sized PM categories of PM_{1.0} and ultrafine particulates in the < 100 nm range (UFP) have been attracting growing research attention (Young et al., 2013; Jayaratne et al., 2015; Krecl et al., 2015; Lin et al., 2015). The PM size is critical to its toxicity by inhalation. It has been reported that UFP can deposit efficiently in the alveolar region upon inhalation, and that the nanoparticles could potentially be taken up by brain cells (Oberdörster et al., 2005: Nel et al., 2006:). It has also been suggested that fine and ultrafine PM (especially from combustion) are more strongly associated with adverse health effects compared to larger particles (Oberdörster et al., 1995; Donaldson et al., 2001; Löndahl et al., 2006; Tsai et al., 2012; Agarwal et al., 2015). A major obstacle to gathering long-term exposure data has been the lack of suitable portable or personal instrumentation that can simultaneously monitor these ultrafine and nanoparticle size distributions (PSDs) in real time (Maynard and Kuempel, 2005; Maynard et al., 2006; Qi and Kulkarni, 2012; Ostraat et al., 2013).

The scanning mobility particle size spectrometer (SMPS)

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system is widely used for measuring aerosol PSDs from 10 nm to 1.0 μ m with high sizing and temporal resolution. The advantages of using the SMPS system are that PSDs can be measured in a relatively short time, and different dose metrics (e.g., the number, surface area, and volume) of PM_{1.0}, ultrafine and nanoparticles can be estimated simultaneously (Cheng et al., 2009; Du et al., 2012; Filep et al., 2013; Fission et al., 2013; Sahu et al., 2013; Stabile et al., 2013; Sarangi et al., 2015). However, the current commercially available SMPS systems (e.g., TSI SMPS 3936 and 3938, USA; GRIMM SMPS+C or SMPS+E, Germany; MSP Wide Range Particle Spectrometer, WPS M1000XP, USA; Palas U-SMPS, Germany) are large, bulky, expensive, and/or use radioactive neutralizers, making them inappropriate for routine exposure monitoring and assessment (Qi and Kulkarni, 2012; Ostraat et al., 2013). A portable and cheap SMPS with high-resolution suitable for the needs of atmospheric monitoring and occupational hygiene is required.

One of the most important developments of a portable SMPS is to design a small differential mobility analyzer (DMA), which are mainly three types of DMA including radial (Hurd and Mullins, 1962; Zhang et al., 1995; Fissan et al., 1996; Qi et al., 2008; Brunelli et al., 2009), cylindrical (Babis et al., 2009; He, 2014) and planar (Erikson, 1921; Knutson and Whitby, 1975; Spangler, 2000; Zimmermann et al., 2007, 2008). The various manufacturers of commercial DMAs utilize similar dimensions (Steer et al., 2014). Both radial and planar geometry DMAs suitable for a portable instrument design can be more effective and simplified than the cylindrical one due to limited space. In addition to classify particle size, the DMA of high resolving power has been used to measure the mobility distribution of air ions generated by radioactive source, ²⁴¹Am and corona discharge as a function of the applied voltage and polarity (Alonso et al., 2009). Recently, two portable particle size distribution analyzers (TSI NanoScan SMPS, 3910, USA and Kanomax PAMS, 3300, Japan) were introduced to the market. These are battery powered, SMPS-type particle sizers that use a Condensation Particle Counter (CPC) for detecting and counting the classified particles. Assessment of the comparability and accuracy of paired portable CPCs of the same type (TSI 3007) with the size range of 10 nm to 1.0 µm shows inconsistency, with a mean difference of 22.9% compared with the laboratory-based SMPS (TSI 3936) (Liu et al., 2014). Metrological assessments have been conducted to characterize the performance of the TSI NanoScan SMPS with respect to the well-established laboratory-based SMPS (Tritscher et al., 2013; Ruths et al., 2014; Stabile et al., 2014). In practical terms, although the size distributions of aerosols in an indoor environment measured by the NanoScan were coincident with the reference SMPS, Yamada et al. (2015) indicated the count concentration of particles with size ranges of 200 to 420 nm measured by the NanoScan was incomparably lower than the reference SMPS, probably due to the countable size being close to the upper size-specific limit. However, to date, no comparison of the TSI NanoScan, the Kanomax PAMS, and the laboratory SMPS has been reported.

The design of a personal/portable mini-SMPS has been

described in detail previously (Qi and Kulkarni, 2012). Briefly, the mini-SMPS system consists of three subsystems: a charger or neutralizer, a differential mobility analyzer (DMA), and a particle counter (using either CPC or an aerosol electrometer, AE). Aerosol particles are first introduced into a bipolar diffusion charger/neutralizer or a unipolar charger where they reach a known and stationary charge distribution in a short time. In the NanoScan, a patented opposed flow "unipolar" diffusion charger is used (Medved et al., 2000), while the PAMS uses a patented miniature dual-corona ionizer as the non-radioactive "bipolar" charger bringing the particles to steady-state charge distribution (Qi and Kulkarni, 2013). In the NanoScan's unipolar diffusion charger, unipolar ions from a corona discharge at a platinum needle tip and the aerosol flow in the opposite direction are swept through subsonic orifices to form turbulent jets. The particles and unipolar ions are then mixed and collided in the mixing chamber. The passive transmission efficiency of the charger used in NanoScan is 80%, and the charging efficiency is around 80% for 50 nm particles. The aerosol passing through the PAMS is bipolar and charged by a dual-corona ionizer (Qi and Kulkarni, 2013). Two identical corona ionizers produce ions of opposite polarities with the same magnitude of corona current, and are positioned in parallel and symmetrically about the aerosol flow axis. The aerosols are exposed to the ions by employing an aerosol flow cavity, and are thus charged. The transmission efficiency is around 80% for particles smaller than 50 nm, and greater than 90% for particles larger than 100 nm. In contrast, the charging efficiency is greater than 90% for 10 nm particles, and decreases as the particle size increases. Both the chargers applied in NanoScan and PAMS are free of radioactive sources, and have the advantages of a small hardware size suitable for field measurements and a low fabrication cost.

After exposure to the chargers, the charged/neutralized particles are then classified using the DMA by selecting different applied voltages. NanoScan utilizes a radial-type DMA (rDMA) (Steer et al., 2014), while the design of the PAMS DMA utilizes a miniature cylindrical DMA, which is operated at the low flow rate of 0.05 L min⁻¹ (lpm) to cover the wide size range in a single geometry. Once the particles have been classified, the number concentration of the aerosol with a very narrow distribution centered at a known electrical mobility, which is a function of the particle size and charge, is determined by the CPC or AE. A CPC enlarges the particles by passing the aerosol stream through the supersaturated vapor of a working liquid before they are counted by the optical sensor, while an AE estimates the particle concentration from measurements of the electric current of charged particles. The NanoScan and the PAMS both utilize the hand-held TSI 3007 CPC (Hämeri et al., 2002; Tritscher et al., 2013), but the sampling flow rates to each device are very different; the NanoScan delivers the aerosol to the CPC at a flow rate of 0.25 lpm, while the PAMS flow rate is 0.05 lpm.

This paper thus examines the performance and monitoring capability of the two portable ultrafine to nanoparticle sizers in terms of particle sizing and counting, and compares them to a reference TSI SMPS 3936, the most commonly used electrical mobility particle spectrometer in aerosol science researches (Rodrigue *et al.*, 2007). The National Institute of Standards and Technology (NIST) also applied a similar system to measure size 60 nm and 100 nm standard size reference materials (Mulholland *et al.*, 2006).

METHODS

A schematic diagram of the operation of the TSI NanoScan 3910 and Kanomax PAMS 3300 is shown in Fig. 1. In operation, both NanoScan and PAMS provide scanning measurements (the scan mode) and one classified-size measurement (the single mode). NanoScan measures the PSD over a size range of 11 nm to 420 nm with a resolution of 13 size bins, and the scanning time is fixed at 1 min. During this time, there is a 45 second up-scan and a 15 second down-scan (retrace). On the other hand, PAMS offers two sizing resolutions (the High Mode and Wide Mode) using different sheath flow rates in its DMA. In the High Mode, the size range is from 10 nm to 433.7 nm, and is divided into 27 size bins. In the Wide Mode, the measuring range is extended to 862.3 nm while the number of size bins is reduced to 14, so as to perform rapid measurements. A detailed comparison of the specifications is shown in Table 1.

Because it is integrated into both the PAMS and NanoScan, the CPC's performance as an independent unit is difficult to assess and this assessment was therefore not made. The performances of the NanoScan and PAMS were compared

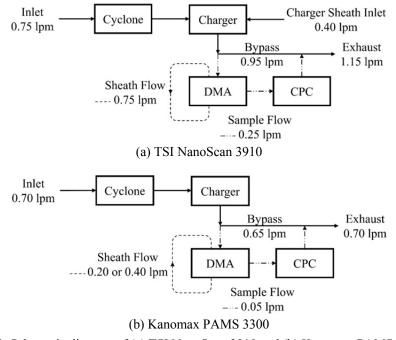


Fig. 1. Schematic diagram of (a) TSI NanoScan 3910 and (b) Kanomax PAMS 3300.

Table 1. A	A comparison of	f the operating	features of	f TSI NanoSc	an 3910 and	Kanomax PAMS3300.

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	TSI NanoScan 3910	Kanomax PAMS 3300			
Flow Rate	Inlet: 0.75 lpm	Inlet: 0.70 lpm			
Flow Rate	Sample: 0.25 lpm	Sample: 0.05 lpm			
	Unipolar charging:	Bipolar charging:			
Charger	Single-corona charger at + 1 mA	Dual-corona charger at $\pm 3 \pm 1 \ \mu A$			
	(Medved et al., 2000)	(Qi and Kulkarni, 2013)			
DMA	rDMA, negative high voltage applied	Cylindrical DMA, negative high voltage			
DIVIA	(Zhang et al., 1995)	applied.			
Measuring Modes	Single Mode and Scan Mode				
	Wide Mode:				
		14.5-863 nm/ 14 channels/			
Scan Resolution	10-420 nm/ 13 channels/ fixed at 60 sec	42 sec-140 min (Sheath: 0.2 lpm)			
Scall Resolution	(Sheath: 0.75 lpm)	<u>High Mode</u> :			
		10-433 nm/ 27 channels/			
		81 sec-270 min (Sheath: 0.4 lpm)			
Concentration Range	$100-1,000,000 \text{ particles cm}^{-3}$	$0-100,000 \text{ particles cm}^{-3}$			
Dimensions ($W \times H \times D$)	$45 \text{ cm} \times 23 \text{ cm} \times 39 \text{ cm}$	$23 \text{ cm} \times 23 \text{ cm} \times 15 \text{ cm}$			
Weight	9 kg (19.5 lbs)	4.9 kg (9.9 lbs)			

to a reference SMPS (TSI 3936) with a long DMA (TSI 3081) and a TSI 3022 CPC. A schematic diagram of the experimental set-up is shown in Fig. 2. Polydispersed sucrose particles were first generated by a collision type atomizer with a dryer and a neutralizer. An Electrostatic Classifier (TSI 3080L) was then applied to classified 100 nm monodispersed sucrose particles for testing. Either NanoScan or PAMS was then chosen with the reference SMPS to perform measurements simultaneously. The reference SMPS was operated at a sheath flow rate of 3.0 lpm and an aerosol flow rate of 0.3 lpm, and the scanning time was set at 60 sec. The experimental set-up with and without an additional Kr⁸⁵ neutralizer (TSI 3077) in front of the PAMS or NanoScan was designed to evaluate the effect of the pre-charged particles on the analyzer performance. Both modes offered by PAMS were tested. The NanoScan uses a unipolar charger while the PAMS applies a bipolar charger. Intrinsically, they have different charging efficiency for the same particle (particles with identical size). In other words, the equilibrium charging state is different for these two devices. The charging tables, of course, are different for their algorithms to invert the particle size distribution. Therefore, in this study, we explore the "overall" effect on their measurements of pre-charged particles. With an additional neutralizer the singly charged particles classified by the DMA are re-neutralized and represent the base reference, while the "overall" effect will be seen without an additional neutralizer. Here, "overall" means that we use the inverted particle size distribution as the end performance parameter. However, the overall effect was a combination of the response of the charger, the transfer function of DMA as well as the soundness of the algorithm.

NanoScan and the reference SMPS were applied to measure the ambient aerosol concentration for one month using an identical sampling inlet. The field measurements were conducted on top of Mt. Lulin (2862 m above sea level; 23.47°N, 120.87°E) located in central Taiwan (Lulin Atmospheric Background Station, LABS, 23.47°N, 120.87°E,) between 5 and 31 March 2014. Mt. Lulin is under the influence of Southeast Asian biomass-burning particle emissions in spring due to transport by high altitude westerlies (Yen *et al.*, 2013).

RESULTS AND DISCUSSION

Comparison of PAMS with the Reference SMPS

The results of the comparison of PAMS with the reference SMPS are shown in Figs. 3 and 4. The major modal sizes of the classified distributions measured by PAMS with and without an additional neutralizer were both located at 96.47 nm, which is slightly smaller than the modal size (98.2 nm) presented by the reference SMPS (Fig. 3). This could be due to the lower sizing resolution for the PAMS compared to the reference SMPS. Based on the measurements from the reference SMPS, the Geometric Standard Deviation (GSD) and the Geometric Mean Diameter (GMD) of the major peak of singly charged aerosols were around 1.10 and 98.13 nm, respectively. The corresponding D_{p.16} and $D_{p,84}$ were 88.8 and 107.45 nm. Within this size range, however, the PAMS only provided one data point at 96.47 nm. even operated at HIGH Mode. On the other hand, the peak concentration of the PAMS measurement without an additional neutralizer $(dN/dlogD_p = 7.74e+5 \# cm^{-3})$ was higher than that obtained with an additional neutralizer $(4.69e+5 \ \text{# cm}^{-3})$ and the reference SMPS measurements $(4.11e+5 \# cm^{-3})$. This was consistently observed for different classified sizes, which are not presented here. In the Wide Mode, the modal sizes of the PSDs measured by PAMS with and without an additional neutralizer were located at 97.5 nm, which was closer to results from the reference SMPS. This was because the available data points were limited by the PAMS software/algorithm, not due to increased sizing accuracy. Moreover, the GSD of the PSDs measured using WIDE Mode was broader and the peak concentration was lower, and the second peak due to doubly charged aerosols was almost unrecognizable and/or smeared (Fig. 4). This can be explained by the influence of the flow ratio on the DMA transfer function (Collins et al., 2004).

The Wide Mode is an extended function offered by PAMS, and allows users to operate the PAMS DMA at a lower sheath flow rate (0.2 lpm) and to have a wider sizing range. The aerosol flow/sample flow is kept constant at 0.05 lpm, but the ratio of the sheath flow to the aerosol flow is reduced from 8:1 to 4:1. Because of the reduction in the sheath to aerosol flow ratio (Q_{sh}/Q_a), the sizing resolution

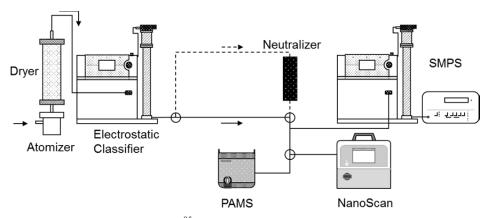


Fig. 2. Experimental set-up with and without a Kr^{85} neutralizer for a comparison of the performance between the reference SMPS and TSI NanoScan or Kanomax PAMS.

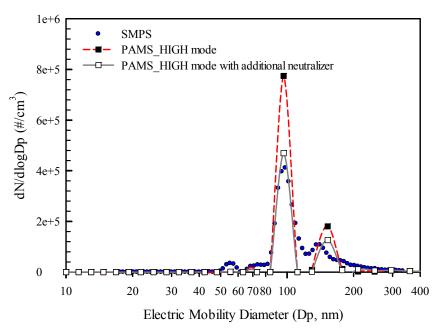


Fig. 3. Measurements of 100 nm monodispersed sucrose particles by SMPS and PAMS under the High Mode with and without an additional neutralizer.

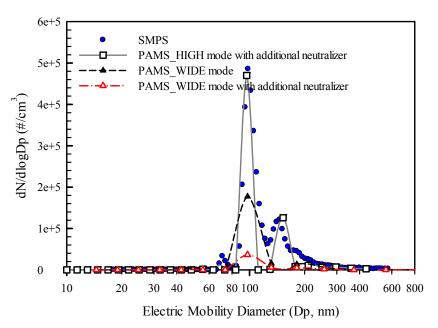


Fig. 4. Measurements of 100 nm monodispersed sucrose particles by SMPS and PAMS under the High Mode and the Wide Mode.

is decreased greatly, as shown in Fig. 4. Therefore, it is a trade-off between wider sizing range and lower resolution here. In addition, the very low sampling flow rate (0.05 lpm) could possibly introduce concentration instability in the CPC, and hence uncertainty in the classification in the DMA of PAMS. Control and stability of the sampling flow rate could therefore be a critical issue for PAMS.

A higher concentration after classification without a neutralizer was also clearly perceived in the PAMS measurements under both the High Mode and Wide Mode. This higher peak concentration may be due to pre-existing charges on the classified particles. As the Electrostatic Classifier applies a negative high voltage on the DMA, classified particles with a similar electric mobility carry positive charges. Although PAMS uses a bipolar corona charger to neutralize these pre-existing charges, the final charge fraction of particles with positive charges would be higher than with an additional neutralizer. Therefore, more particles pass through the DMA of PAMS (with applied negative high voltages) than expected, and the inverted concentration is thus higher. The effects of pre-existing charges on unipolar charging and electrical aerosol measurements were discussed thoroughly in Qi's work (Qi *et al.*, 2009). This effect could be more significant for NanoScan, as it utilizes a unipolar corona charger. However, from the experimental results, Fig. 5 shows very minor differences between the cases with and without an additional neutralizer for NanoScan. A more detailed performance characterization and comparison of the chargers in NanoScan and PAMS chargers are needed.

Comparison of NanoScan with the Reference SMPS

In contrast, the sampling flow rate of NanoScan (0.25 lpm) is much higher than that of PAMS. Conversely, the sheath to aerosol flow ratio (Q_{sh}/Q_a) in NanoScan is constant

at 5:1, which is close to the value for the Wide Mode of PAMS. Therefore, compared to the reference SMPS and PAMS under the HIGH Mode, both NanoScan and PAMS under the Wide Mode would give a lower resolution when measuring monodispersed PSD. Note that while PAMS in the Wide Mode and NanoScan provide a similar number of size bins, NanoScan measures particle sizes only up to 420 nm. Although we do not know the design of the DMA of PAMS and cannot conduct a detailed analysis of the performance of their DMAs, both NanoScan and PAMS measurements under the Wide Mode share the similarity of broader PSD and lower peak concentration and therefore the lower Q_{sh}/Q_a value in NanoScan could be one of the reasons

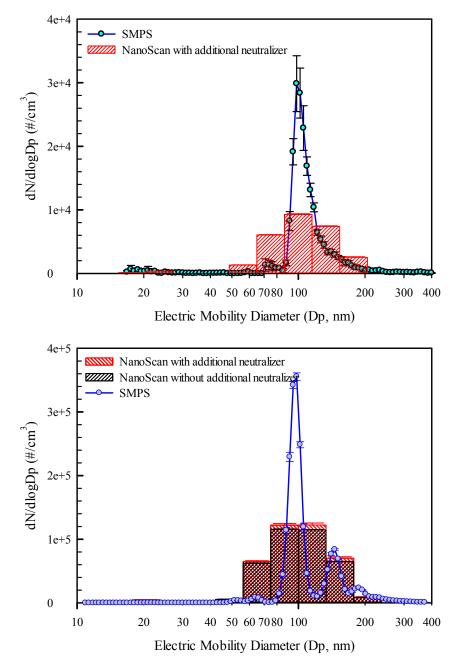


Fig. 5. Measurements of 100 nm monodispersed sucrose particles by SMPS and NanoScan with and without an additional neutralizer.

for the coarser resolution when measuring monodispersed particles (Fig. 5). This was also shown in a study by Tritscher et al. (2013). On the other hand, the total concentrations measured by the reference SMPS and the NanoScan differ by only about 5~8%, which is within the CPC performance variation suggested by the manufacturer, TSI. In other words, if the resolution of the PSD presented by the reference SMPS was reduced to the same level as the NanoScan $(dlog D_p)$ ~0.125), their PSDs would look similar. This indirectly implies that the broadened and smeared PSD was due to the coarser resolution. In addition, we also generated the bi-modal PSD (with the second doubly charged aerosols) for testing the size resolution. As seen in the lower plot of Fig. 5, the second peak was almost unrecognizable, similar to the PAMS under WIDE Mode. For ambient polydispersed measurements, shown in Fig. 6, NanoScan and the reference SMPS showed good agreement with high correlation coefficient (r value) of 0.927, but deviation was observed for particles with sizes of less than 30 nm. This could be due to the different counting efficiency of the CPC used in NanoScan and the reference SMPS, and the insufficient charging efficiency and/or non-spherical morphology of the particles in the lower size range. In practical terms, although the size distributions of the real aerosols in indoor environment measured by the NanoScan were coincident with the reference SMPS, Yamada et al. (2015) indicated the count concentration of particles with size ranges of 200 to 420 nm measured by the NanoScan was incomparably lower than the reference SMPS, probably due to the countable size being close to the upper size-specific limit.

Practical Comparison of NanoScan and CPC with the Reference SMPS

In previous investigations, Tritscher *et al.* (2013) and Ruths *et al.* (2014) both reported good comparability of NanoScan to the TSI 3936 SMPS in measuring particle concentrations. Tritscher et al. (2013) generated challenging particles in the lab, and concentration linearity was maintained up to 10^6 particles cm⁻³. In contrast, Ruths *et al.* (2014) conducted their experiements in the field, and showed that deviations between NanoScan and the reference SMPS were 10% to 15%. Stabile et al. (2014) found that NanoScan overestimated the total particle concentration for aggregate particles, such as fresh diesel emissions, and attributed this to pre-existing charges and the unipolar charger in NanoScan. Based on our one-month long field measurements at LABS in central Taiwan in the spring of 2014, the total particle number concentrations measured by NanoScan and the Optical Particle Sizer (OPS, TSI 3330) compare moderately well with the measurements of the reference SMPS and those of a separate TSI 3010 CPC (Fig. 7). The scatter plots with linear regressions of the total number concentrations measured by the CPC 3010, SMPS and NanoScan with OPS are shown in Fig. 8. The regression coefficient of 0.619 between SMPS, and NanoScan and OPS was moderate, whereas that between CPC, and NanoScan and OPS (0.941) was high. Although the trends of total particle number concentrations measured by the three analyzers are similar to each other, overestimation or underestimation was seldom observed, and the deviations were in the order of about 19%.

CONCLUSIONS

There are two critical investigable differences between NanoScan and PAMS: the different sheath to aerosol flow ratio (Q_{sh}/Q_a) and the different charging techniques. The different DMA designs in these two portable sizers could be important as well. However, the specifications of the PAMS DMA are not available in either the manufacturer's

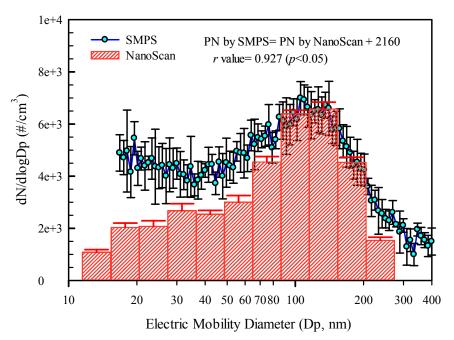


Fig. 6. Measurements of polydispersed particles by SMPS and NanoScan.

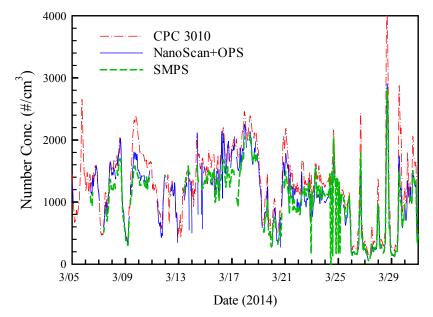


Fig. 7. Comparison of the total particle number concentration measured by CPC 3010, SMPS, and NanoScan with OPS.

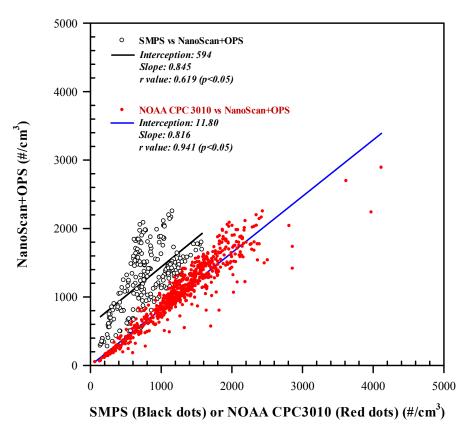


Fig. 8. Scatter plots with linear regressions of the total particle number concentrations measured by CPC 3010, SMPS and NanoScan with OPS.

manual or the reported literature. Therefore, only the sheath to aerosol flow ratio (Q_{sh}/Q_a) was investigated in this study. Regarding Q_{sh}/Q_a , PAMS provides two operational modes corresponding to two Q_{sh}/Q_a ratios (8:1 and 4:1) in its DMA, while NanoScan has only one fixed Q_{sh}/Q_a ratio of 5:1. When PAMS was operated at the high Q_{sh}/Q_a ratio

(High Mode), the sizing resolution was greatly improved. Therefore, PAMS can offer almost double the number of size bins of NanoScan within a similar measuring size range. However, the very low sampling flow rate (0.05 lpm) of PAMS can cause uncertainty in the classification process inside the DMA and in the concentration measurements in the integrated CPC. The stability of this very low flow rate is therefore a critical issue for PAMS. The stable control of such a low flow rate in a compact design is an engineering challenge, and further testing should be conducted for long-run operations.

NanoScan uses a mixing type unipolar charger with positive ions, which could be sensitive to pre-existing charges on the particles. Stabile et al. (2014) have suggested remedying this by having a separate CPC in tandem with NanoScan to verify and correct the measured PSD. However, based on our experimental results on measuring monodispersed PSD, the performance of NanoScan was not significantly influenced by pre-existing charges on the particles. PAMS uses a dual-needle bipolar corona charger to neutralize incoming particles. The effect of pre-existing charges was thus expected to be alleviated. However, it was observed that the measured concentration of particles with pre-existing positive charges was higher than that of neutralized particles. In other words, the over-counting of aggregate particles reported by Stabile et al. (2014) could be an issue for PAMS as well.

For measurements of polydispersed particles, both NanoScan and PAMS showed comparable capability for near real-time particle distribution measurement. Based on the one-month field measurements at the Lulin Atmosphere Background Station (LABS) in Taiwan in the spring of 2014, the average deviation between NanoScan and the reference SMPS was about 19%. For measurements of monodispersed particles, PAMS offers a higher sizing resolution under High Mode operation than NanoScan does. On the other hand, NanoScan allows for the possibility of combining measurements with the TSI OPS, and this could extend the overall sizing range by up to 10 μm.

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