

Development of a new Nano-particle sizer equipped with a 12 channel multi-port differential mobility analyzer and multi-condensation particle counters

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Abstract

Measuring particle size distributions precisely is an important concern in addressing environmental and human health-related issues. To measure particle size distributions, a scanning mobility particle sizer (SMPS) is often used. However, it is difficult to analyze particle size distributions under fast-changing concentrations conditions because the SMPS cannot respond fast enough to reflect current conditions due to the time necessary for voltage scanning. In this research, we developed a new Nano-particle sizer (NPS), which consists of a multi-port differential mobility analyzer (MP-DMA) with 12 sampling ports and multi-condensation particle counters (M-CPCs) that simultaneously measure concentrations of particles classified by the sampling ports. The M-CPC can completely condense particles larger than 10 nm, and the total particle concentrations measured by each homemade CPC in the M-CPCs and an electrometer were in agreement up to 20,000 # cm⁻³. We conducted size distribution measurements under steady-state conditions using an aerosol generator and under unsteady conditions by switching the aerosol supply on/off. The data obtained by the NPS corresponded closely with the SMPS measurement data for the steady-state particle concentration case. In addition, the NPS could successfully capture the changes in particle size distribution under fast-changing particle concentration conditions. Finally, we present NPS measurement results of size distributions in common situation (cooking) as an exemplary real-world application.

Keywords: Nano-particle sizer; scanning mobility particle sizer; multi-port differential mobility analyzer; multi-condensation particle counter; real-time particle size distribution; unsteady particle size distribution

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1 Introduction

There are several methods to measure size distributions of aerosols. Among them, the combination of a differential mobility analyzer (DMA) and a condensation particle counter (CPC) has been widely used. The measurement procedure of this technique begins with a voltage applied to the DMA to classify monodisperse particles in a narrow electrical mobility range, and then the CPC measures the particle number concentration (Fissan et al., 1983). This is the differential mobility particle sizer (DMPS) method, and by stepping the voltages, the complete size distribution of aerosols can be obtained. However, generally 10–15 min of the voltage stepping process are required for accurate estimation of the complete size distribution, making the DMPS unable to respond accurately if the concentration is changing rapidly. For this reason, the DMPS method has limited applications. Wang and Flagan (1990) developed a scanning mobility particle sizer (SMPS) to reduce the measurement time. For the SMPS measurement, the applied voltage is increased (or decreased) continuously, and particles consecutively classified by a DMA are counted by a CPC. As a result, measurement time can be reduced to less than two minutes. However, it is still too long to analyze fast-changing particle size distributions. Recently, several aerosol instrument systems have been developed and studied with the aim of faster measurement. A fast mobility particle sizer (FMPS) was developed based on a principle similar to the SMPS system, the electrical mobility analyzer. Instead of a CPC, the FMPS uses multiple electrometers for particle detection, and the system provides particle size distribution information in real time. The FMPS is generally used for analyzing engine emissions because the electrometers are not sensitive enough to measure low particle concentrations ($< 10^2 \text{ \# cm}^{-3}$). In addition, current leakage and electrical noise of electrometers sometimes result in less precise measurements. A new fast integrated mobility spectrometer (FIMS) for real-time measurement of aerosols was developed (Kulkarni and Wang, 2006). The FIMS detects charged particles based on their different electrical mobilities, which result in different trajectories. A fast charge-coupled device (CCD) imaging system is employed to capture the locations of droplets nucleated from these spatially separated particles. The FIMS can be used to obtain size distributions at sub-second time intervals. Another fast aerosol measurement instrument is a DMA-train (Stolzenburg et al., 2017). The DMA-train is operated with six DMAs in parallel at a fixed voltage for particle size distribution measurement with high-time resolution. Therefore, it can be used to observe very fast aerosol growth, especially in the sub-10 nm range. However, the DMA-train contains six commercial CPCs and six commercial DMAs, which make the system costly and bulky. Recently, Oberreit et al. (2014) performed mobility analysis of sub-10 nm particles using an aspirating drift tube ion mobility spectrometer (DT-IMS) numerically and experimentally.

By using the instrument, the electrical mobility of the particles can be estimated from the time required for the particles to traverse a drift zone. The findings in the paper show that particles ranging from 2 to 11 nm can be analyzed in less than 5 s. Another instrument for fast measurement is a nucleation mode aerosol size spectrometer (NMASS) developed by Williamson et al. (2018). The NMASS consists of five embedded CPCs with different cut-off diameters to measure the particle size distribution between 3 and 60 nm. To distinguish different diameters, the NMASS requires five different thermal operating conditions for its condensers.

In addition to the above-mentioned instruments, Chen et al. (2007) and Kim et al. (2007) previously developed a differential mobility analyzer with multiple sampling ports for a fast measurement system. However, the multi-stage DMA (MDMA) by Chen et al. (2007) has only three sampling ports and needs three CPCs. Furthermore, an exponentially extended longitudinal length is required to increase the number of sampling ports and accommodate the wide size range of particles. As a result, the system becomes complicated and expensive. Kim et al. (2007) developed a DMA with a multi-port system, a substitution for the MDMA system, and it can classify a total of seven sizes simultaneously. They evaluated the DMA system using monodisperse particles and deduced from the experiments that increasing the number of sampling ports did not affect the classification efficiency and transfer functions of the DMA. This was also theoretically supported in research by Giamarelou et al. (2012), in deriving analytical expressions for estimating the transfer functions and the resolutions of DMAs with multiple sampling ports. However, there is still a lack of research on a fast measurement system that retains the traditional DMA function. Therefore, in this study, we developed a new Nano-particle sizer (NPS), consisting of a multi-port DMA (MP-DMA) and multi-CPCs (M-CPCs), that can perform fast measurement of particle size distributions.

2 Instrument

2.1 Design Concept and Construction of the NPS

The NPS consists of one MP-DMA with 12 ports (Fig. 1(a)) and two M-CPC modules with 12 homemade CPCs (Fig. 1(c)). The MP-DMA, unlike the common cylindrical DMA with one sampling port (Knutson and Whitby, 1975), has an outer electrode with multiple sampling ports (annular slits) and a truncated cone-shaped inner electrode where a high voltage is applied. Once the constant voltage is applied, the MP-DMA classifies monodisperse particles according to their electrical mobility. The dimensions of the entire system are $450 \times 300 \times 250$ mm. The flow systems and paths for the NPS are depicted in Fig. 1, including the aerosol flowrate (Q_a , 0.18 L min⁻¹), sheath flowrate (Q_{sh} , 3.78 L min⁻¹), sampling flowrate for each CPC (Q_s , 0.18 L min⁻¹), and exhaust

flowrate (Q_e , 1.8 L min^{-1}). Like the common DMA flow system, Q_a is the same as Q_s . The clean sheath flow carries aerosols from the top to the bottom. Because Q_s continuously flows out through each sampling port, the total flowrate along the classification zone is reduced.

2.2 Design Concept of the MP-DMA

While Chen et al. (2007) employed three sampling ports and applied an exponentially increasing distance between neighboring ports to allow a wide size range of particles, the MP-DMA has 12 annular sampling ports that are placed with a uniform distance of 2 cm between neighboring ports. The MP-DMA uses an inner electrode with increasing diameter along the longitudinal direction. As the diameter of the electrode increases, the distance between the inner electrode and the outer cylindrical electrode decreases. Accordingly, the electrical field strength applied to particles increases as they flow to the downstream side. As a result, the MP-DMA can accommodate a wider size range of particles without excessive extension of the electrode length found in the common cylindrical electrode.

2.3 Design Concept of the M-CPC

Each sampling port in the MP-DMA is directly connected to the inlet of each homemade CPC. Classified particles are introduced to and measured by the CPC. One M-CPC module consists of six homemade CPCs, and the NPS has two M-CPC modules (12 CPCs). The module has a unified saturator and condenser block to maintain uniform temperatures. A common working fluid reservoir is located beneath the saturator block. The operating principle of the M-CPC is same as other typical CPCs. Particles are introduced to the saturator (temperature: 35°C), and the condensational growth of the particles occurs in the condenser at a temperature of 10°C . The condensed particles are detected in the optical part. Each homemade CPC was denoted as CPC1, CPC2, CPC3, etc., based on their location. CPC1 is closest to the aerosol inlet and CPC12 is closest to the sheath outlet in the MP-DMA. The reference CPC used in this study is denoted as TSI-CPC (model 3776, TSI Inc., Shoreview MN, USA).

Figure 1

3 Experimental Setup and Operating Conditions

3.1 M-CPC

In order to evaluate the performance of the M-CPC, the activation efficiency and concentration linearity of each

homemade CPC were obtained from comparison with a reference electrometer. Figure 2(a) is the schematic diagram of the M-CPC performance test. Using a homemade Collison atomizer, a 0.1 wt% NaCl solution was atomized, and the aerosols were classified by the first DMA (standard DMA, model 3081, TSI Inc., Shoreview MN, USA) to generate monodisperse particles which were distributed to the analyzing instruments. In this study, the operating sheath and aerosol flowrates in the first DMA were 10 L min^{-1} and 1 L min^{-1} , respectively. The mode size and geometric standard deviation of the atomized aerosols were 43 nm and 1.65, respectively. The particle sizes obtained from the atomizer were smaller than 100 nm, thereby minimizing multiple charging effects on the size-selection (Fig. S1 in Supplementary Material). The concentration of particles was controlled by a diluter before entering the instruments as shown in Fig. 2(a). To measure the particle number concentration as a reference, an electrometer (model 6517A, Keithley) with a Faraday cup was used. This is one of the most commonly used methods for CPC calibration (Liu and Pui, 1974). In this experiment, the sampling flowrate of each CPC was 0.18 L min^{-1} , and N-butyl alcohol (Agarwal and Sem, 1980) was used for the working fluid. Temperatures of the condenser and saturator were controlled to maintain 10°C and 35°C , respectively. The M-CPC measured the number concentration every 1 s, and the response time of the M-CPC is less than 0.3 s. The experimental setup shown in Fig. 2(a) was used to obtain the results in Fig. 3. For the activation efficiency tests, the tested particle sizes were 10 nm, 30 nm, 50 nm, 80 nm, and 100 nm. For the concentration linearity test, which is associated with the detection efficiency of M-CPCs, 50 nm monodisperse particles were used. The tested monodisperse particles were introduced to the sheath inlet of the MP-DMA with 0 V applied to the inner electrode, and the concentrations measured by each CPC and the electrometer were compared.

3.2 MP-DMA

To evaluate the performance of the MP-DMA, the normalized particle mobility distribution for each port and penetration efficiency for the MP-DMA were obtained. Figure 2(b) is the schematic diagram of the MP-DMA performance test. The particle size and concentration were controlled by the first DMA and diluter, respectively. The operating conditions of the MP-DMA were 0.18 L min^{-1} for Q_a , 0.18 L min^{-1} for Q_s , 1.8 L min^{-1} for Q_e , and 3.78 L min^{-1} for Q_{sh} . The total flowrate ($Q_{sh} + Q_a$) flowing inside the MP-DMA decreases as the flow goes along the downstream side because each CPC takes 0.18 L min^{-1} . Under these flow conditions, the residence time of the particles flowing from the aerosol inlet to each sampling port inlet is approximately 0.3 s (Port 1) to 3 s (Port 12) (Lee et al., 2020). The delay due to the residence time inside the MP-DMA was considered when obtaining the size distributions. In the experiments, the applied voltage on the MP-DMA was fixed, and the stepwise increase

of the voltage on the first DMA was carried out to generate different sizes of monodisperse particles. Their concentrations were measured by each CPC in the M-CPCs. The upstream concentration of the monodisperse particles was monitored by the reference TSI-CPC and controlled to approximately $10,000 \text{ \# cm}^{-3}$ by adjusting the valve ('B' in Fig. 2(b)) located in the diluter.

With step-wise increase of the voltage on the first DMA, the mobility distributions were obtained from the sets of measured concentrations as a function of electrical mobility based on the first DMA. The measured concentrations were normalized by the maximum concentration for each port. The electrical mobility was normalized by the central mobility for each port, and the results are shown in Fig. 4. In addition, the particle penetration ratios as a function of port number at voltages of 1000 V and 2000 V are shown in Fig. 5, representing the maximum ratio between the measured concentration at each CPC and the upstream concentration measured by the TSI-CPC, which is approximately $10,000 \text{ \# cm}^{-3}$. The maximum penetration ratio was obtained at the central electrical mobility for each port. The penetration ratios were used to calibrate the NPS data in the inversion process.

3.3 Particle Size Distribution Measurement

To test the performance of the NPS, the experimental set-up in Fig. 2(c) was used. For particle generation, we used two types of particles, NaCl and Ag. The NaCl and Ag particles were generated by the homemade Collision atomizer and evaporation generator (Hwang and Ahn, 2017). The particles were neutralized by a neutralizer, and the concentration was controlled by a diluter. The particles were introduced into the TSI-SMPS and NPS. The TSI-SMPS consists of the standard long DMA (model 3081, TSI Inc., Shoreview MN, USA) and a CPC (model 3775, TSI Inc., Shoreview MN, USA), and the voltage was generated by a high-voltage power supply (model 205B-10R, Bertan High Voltage, Hicksville NY, USA). The NPS was operated at a constant voltage of 1000 V for size distribution measurements. The performance tests were conducted under steady-state conditions with constant NaCl and Ag particle concentrations and with changing NaCl particle concentrations during the transition to the equilibrium state. To provide unsteady particle concentrations, we used the on/off valve at the aerosol path ('A' in Fig. 2(c)) before the TSI-SMPS and NPS. The total measurement time was 240 s. Two cycles of the TSI-SMPS measurement were performed consecutively with 120 s scanning time for each cycle, and the NPS obtained concentration data every 1 s.

Figure 2

3.4 Inversion Process for the NPS Concentration Data

The raw concentration data measured by the M-CPCs were converted to the real concentrations using an inversion process considering the multiple charging effect, detection efficiency of the M-CPCs, and penetration ratio through the MP-DMA. The real concentration of each sampling port was estimated by Eq. (1), and the multiple charge correction was referred by Hoppel's inversion method (Hoppel, 1978). Variables used in this inversion process were derived from the experimental results and research of Giamarelou et al. (2012) and Stolzenburg and McMurry (2008). The correction based on the charge fraction was referred by Wiedensohler's bipolar charge distribution (Wiedensohler, 1988). For a clear understanding of the variables in Eq. (1), we added a brief explanation of the experimental method in each result section.

$$\left. \frac{dN}{d \log D_p} \right|_{D_p^*} = \frac{2 \times N_{raw}(D_p^*)_n \times (60/1000)}{f_c(D_p^*)_n \times P(D_p^*)_n \times \eta_{CPC,act}(D_p^*)_n \times \eta_{CPC,det}(N_{raw})_n \times \{\log(D_{p,E})_n - \log(D_{p,S})_n\}} \quad (1)$$

where D_p is the particle diameter, f_c is the charge fraction, P is the penetration ratio, and $\eta_{CPC,act}$ and $\eta_{CPC,det}$ are the activation and detection efficiency of the M-CPC, respectively. The subscript 'n' indicates the port number. $D_{p,S}$ and $D_{p,E}$ indicate the particle size range classified by each port. Because the NPS receives data every 1 s, the raw data with a unit of # s⁻¹ were converted to # cm⁻³.

4 Result and Discussion

4.1 Performance of the M-CPC

Figure 3(a) shows the activation efficiency of the M-CPCs for particles sizes between 10 nm and 100 nm. To obtain the activation efficiency, monodisperse particles were measured by the electrometer and NPS operated at 0 V as shown in Fig. 2(a). For the NPS measurement, all aerosols were introduced through the sheath flow inlet only (with a flowrate of 3.96 L min⁻¹), so the particle concentrations could be measured by all M-CPCs. The same flowrate of 3.96 L min⁻¹ was introduced to the electrometer, and the measurements were carried out simultaneously. When comparing the M-CPCs to the electrometer measurements, activation efficiencies of almost 100 % were obtained for all CPCs for particle sizes down to 10 nm. In this study, we did not find the cut-size of the M-CPC, but we initially designed the NPS system for detecting particles down to 10 nm.

We also examined the detectable concentration range for the M-CPCs using the experimental setup in Fig. 2(a). The test was conducted with 50 nm monodisperse particles under different concentration conditions. The comparison between concentrations obtained by the electrometer and the M-CPCs is shown in Fig. 3(b). The slope of the graph has a good linearity for concentrations up to $20,000 \text{ \# cm}^{-3}$, indicating that each homemade CPC can be used for concentrations up to this value. It should be noted that a correction factor was considered in the concentration range higher than $20,000 \text{ \# cm}^{-3}$. Furthermore, each CPC in the NPS always measures the concentration of particles classified by the MP-DMA; therefore, in real applications such as atmospheric particle measurements, this high concentration after classified by the MP-DMA can be rarely achieved.

Figure 3

4.2 Performance of the MP-DMA

The normalized mobility distributions of the MP-DMA's 12 sampling ports were obtained using the experimental setup in Fig. 2(b), and the results are shown in Fig. 4. The geometric standard deviations for the distributions were estimated between 1.037 and 1.066, which can be considered a very narrow size classification, indicating that the resolution of the MP-DMA is fairly good. As mentioned earlier, the total flowrate inside the MP-DMA decreases as it flows along the downstream side due to the individual sampling ports continuously taking 0.18 L min^{-1} . Thus, the increase in the ratio of Q_a to Q_{sh} results in increasing geometric standard deviation with increasing port number.

Figure 4

Figure 5 shows the penetration ratio of each port in the MP-DMA at voltages of 1000 V and 2000 V. The penetration ratio is defined as the ratio of the total concentration at the central particle diameter (ref. Table 1) measured by the NPS to the reference concentration obtained by the TSI-CPC as presented in Fig. 2(b). For example, monodisperse particles with a mode diameter shown in Table 1 were generated by using a DMA and introduced to the NPS and TSI-CPC to achieve the penetration ratio. The penetration ratio of the MP-DMA ranges from 0.099 to 0.765, and these data were used for calibrating the NPS system to convert the raw data obtained by the NPS to the reference concentration data. The theoretical resolution of the MP-DMA decreases from 21 (Port 1) to 10 (Port 12) due to the increasing aerosol-to-sheath flowrate. However, the resolution of the first DMA (TSI standard DMA) is 10 owing to the ratio between aerosol and sheath flowrate of 1:10. Therefore, the CPC at Port 1 might count the particles in the narrower size distribution classified by the first DMA, resulting in a low penetration ratio. Thus, the penetration ratios for all ports were used as correction factors in Eq. (1) to achieve the

same concentration as the reference data measured by the TSI-CPC. Notably, in this experiment, the reference data are the concentrations of particles classified by the first DMA, and thus the shape of the input particle size distribution is close to a triangle. Therefore, N_{raw}/P (measured raw concentration divided by the penetration ratio) represents the area under a triangle. For this reason we multiplied a factor of 2 as shown in Eq. (1) assuming that a shape of the size distribution of particles entering each port in the NPS is rectangular.

Figure 5

Table 1 summarizes the central particle diameters on each port under different voltage conditions, 1000 V and 2000 V. The classified mode diameter is the corresponding particle diameter when the concentration of the classified particles in each port is at its maximum. The classified size range of the NPS is 17–210 nm at 1000 V and 25–320 nm at 2000 V. The range can be easily adjusted by changing the applied voltage of the NPS. However, there still remains a limitation in the MP-DMA. There is a blank area between Port 1 and Port 2 where particles with a geometric standard deviation less than 1.04 (narrow size distribution) and a mode diameter between those of Port 1 and Port 2 are deposited and will not be detected. However, most real-world aerosol systems have a wide range of size distribution. Furthermore, the size distribution of aerosols with a geometric standard deviation of 1.04 is rarely seen in actual applications such as a measurement in ambient air. Therefore, the limitation on the MP-DMA might not result in critical issues for atmospheric research purposes.

Table 1

4.3 Performance of the NPS

4.3.1 Steady-state particle size distribution

Using the experimental setup in Fig. 2(c), we introduced NaCl or Ag particles to the NPS to measure particle size distribution, and the results were compared to the TSI-SMPS measurements. The TSI-SMPS system consists of the TSI standard DMA and TSI-CPC which were used in Fig. 2(a) or 2(b). The initial concentrations measured by the NPS were converted to the real concentration based on the inversion process using Eq. (1). Figure 6 shows particle size distributions estimated by the TSI-SMPS and NPS under steady-state conditions of an aerosol generator. The data points from the NPS measurements agree with the TSI-SMPS data. Because the NPS has 12 sampling ports and is operated at a fixed voltage, the number of data points is 12. Therefore, to get the complete size distribution, we fitted the measured data points based on a log-normal distribution. To validate the accuracy

of the fitting method used in this study, we also measured different sets of polydisperse particles (total of ten size distributions) using the TSI-SMPS and NPS to obtain the mode size and total concentration of each size distribution, represented in Fig. 7(a) and 7(b). Overall, the NPS shows comparable performance to the TSI-SMPS in measuring particle size and total concentration, and thus, size distribution. For all TSI-SMPS measurements performed in this study, the corrections for the multiple charging and diffusion loss were applied.

Figure 6

Figure 7

4.3.2 Unsteady particle size distribution

By using the same experimental setup shown in Fig. 2(c), we conducted performance tests for the NPS for unsteady particle size distributions by employing an on/off valve ('A' in Fig. 2(c)) to introduce or block aerosols to the instruments. Fig. 8 shows the comparison between the particle size distributions obtained by the TSI-SMPS and NPS for 240-s measurements. The dotted red line in Fig. 8(a-3) represents the moment we opened the valve ('A' in Fig. 2(c)), indicating introduction of aerosols 60 s after the beginning of the first TSI-SMPS (or NPS) measurement. In Fig. 8(b), we closed the valve to block the aerosols 60 s after the second TSI-SMPS measurement start (i.e., 180 s after the NPS measurement start). The x-axis and y-axis of the graph for the TSI-SMPS measurement results are particle diameter and number concentration, respectively. The NPS data is represented in a contour graph with the sampling time (x-axis) and particle diameter (y-axis). The color indicates the particle number concentration measured by the NPS.

In Fig. 8(a-1), the concentration data appeared after the valve was opened (60 s after the first SMPS scan began). However, concentrations for particle sizes <32 nm were not recovered from the inversion of this scan because the corresponding voltages were applied to the DMA before the valve was opened, when there were no particles in the sample line. In the second scan, the complete size distribution was obtained. In contrast, the NPS measurement shows a rapid increase in particle concentration for the complete size range soon after the valve was opened. Specifically, the particle concentration started to increase or decrease approximately 5 s after the valve was opened or closed, respectively (Fig. S2 in Supplementary Material). Considering the response time of the NPS is approximately 3.3 s, (sum of the M-CPC response time of approximately 0.3 s and particle residence time in the MP-DMA, maximum 3 s), the rest of the delay time might be caused by the time required for concentration stabilization and particle transport. During the test for rapid decrease in particle concentration (Fig. 8(b)), the

performances of the TSI-SMPS and NPS were quite distinct as well. After closing the aerosol valve ~180 s after the measurement, data from the second scan of the TSI-SMPS showed only smaller particles, in a manner similar to the results in Fig. 8(a). However, the size distribution measured by the NPS quickly approached zero. These tests indicate that the NPS can be successfully used for unsteady particle size distributions to observe changes in concentration.

Figure 8

Further NPS measurements under unsteady conditions of rapid-changing particle concentrations were performed for real-world applications. Figure 9(a) and 9(b) represent particle size distributions measured by TSI-SMPS and NPS, respectively, during the cooking of fish. The sampling location for the TSI-SMPS and NPS measurements was 1 m away from the cooking spot, which caused sudden changes in concentration. The cooking activity was continued for approximately 8 min. The size distribution obtained by the NPS is shown every 1 s while the TSI-SMPS measurement provides one size distribution every 2 min (total 6 successive measurements). Therefore, the SMPS analysis provides only discontinuous size distributions. Figure 9(c) shows the particle concentration at a peak particle size for each measurement of the TSI-SMPS and NPS. From the NPS measurements during the cooking activity, particle concentrations varied significantly. Relatively low particle concentrations were observed approximately 180 s after the beginning of the activity, and then several peaks were observed until the end of the event. Like these experiments, size distribution data obtained every 1 s by the NPS can be informative in various applications.

Figure 9

5 Conclusion

We developed and evaluated the performance of a new Nano-particle sizer (NPS) that measures particle size distributions under unsteady conditions with changing concentrations. The NPS consists of a multiport-differential mobility analyzer (MP-DMA) that classifies 12 monodisperse particles of different size and multi-condensation particle counters (M-CPCs) that count the classified particles. The performances of the MP-DMA and M-CPC were evaluated by obtaining activation efficiency, detection efficiency, penetration ratio, and normalized size distributions. The results were used to calibrate the NPS raw data to derive a real particle number concentration and size distribution. The NPS was compared to a TSI-scanning mobility particle sizer (TSI-SMPS) for steady-state and unsteady particle concentrations using NaCl and Ag particles. The size distributions obtained by the NPS

under steady-state condition agreed with the results from the TSI-SMPS. For unsteady particle size distributions with fast-changing particle concentrations, the NPS was found to be superior to the TSI-SMPS in terms of measurement speed. However, there remains a needed improvement. During the NPS measurements, we experienced electrical breakdown when the applied voltage was approximately 4000–5000 V. Therefore, to improve the NPS system for wider size range classification, further optimization is required. From the findings in this study, we believe that the NPS is a promising instrument for providing comprehensive information on particle size distributions in fast-changing concentration environments.

Author contribution

Kang-Ho Ahn conceptualized the instrument developed in this study (Nano-particle sizer), and he received the fund for supporting the project leading to the development of the instrument. Hong Ku Lee designed the experimental methodology and collected the data, and he wrote the original draft. Handol Lee validated and analyzed the experimental data. He wrote the final draft and edited the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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373 Table 1. Mode diameter of the size distribution obtained by using the central mobility range for each port.

	MP-DMA voltage	Port number											
		1	2	3	4	5	6	7	8	9	10	11	12
Mode diameter [nm]	1000 V	17.4	28.9	38.8	48.8	59.9	71.8	84.3	98.6	120.7	138.0	167.2	206.2
	2000 V	24.4	40.7	54.9	70.1	86.6	103.8	122.9	145.5	177.6	207.9	254.5	315.6

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Captions

Figure 1. Schematic diagram of the NPS consisting of the MP-DMA including M-CPCs: (a) the geometry of the MP-DMA and flow paths; (b) the details of the 12th home-made CPC; (c) the M-CPC module.

Figure 2. Schematic diagrams of (a) the M-CPC, (b) MP-DMA and (c) NPS performance tests.

Figure 3. M-CPC performance: (a) activation efficiencies of 12 home-made CPCs; (b) concentration linearity between the electrometer and M-CPCs.

Figure 4. Normalized concentrations of the classified particles through each port in the MP-DMA as a function of normalized electrical mobilities. The C and C^* in the y-axis represent the concentration and the maximum concentration at each port measured by each home-made CPC, respectively. The data were obtained at the NPS applied voltage of 1000 V.

Figure 5. Penetration ratio for each port in the MP-DMA.

Figure 6. Size distributions of the TSI-SMPS and NPS for the constant particle concentrations: (a) Ag particle: evaporation generator (low temperature), (b) Ag particle: evaporation generator (high temperature), (c) NaCl particle: Collision atomizer (0.1 wt% NaCl solution). The data were obtained at the NPS applied voltage of 1000 V.

Figure 7. Comparison of (a) mode sizes and (b) total particle number concentrations obtained by the TSI-SMPS and NPS with NaCl particles. The data were obtained at the NPS applied voltage of 1000 V.

Figure 8. Comparison of the size distributions measured by the TSI-SMPS and NPS for the unsteady particle size distribution in (a) increasing and (b) decreasing particle concentrations. The tested aerosols were introduced or blocked 60 s or 180 s after starting measurements, respectively: (1) the first TSI-SMPS scanning data; (2) the second TSI-SMPS scanning data; (3) the NPS data for 240 s. The data were obtained at the NPS applied voltage of 1000 V.

Figure 9. Size distributions measured by the (a) TSI-SMPS and (b) NPS during a cooking activity and (c) variation of particle concentration at mode diameters. The NPS data were obtained at the applied voltage of 1000 V.

(b)

Diagram illustrating the experimental setup for the study of the thermodynamic cycle of a single droplet. The setup includes a vertical chamber containing a droplet, surrounded by various components: Insulator, Porous, Heater, Cooler, Electrode, and Working Fluid. The chamber is connected to a Reservoir & Drain at the bottom. A vertical arrow labeled Q_s indicates the direction of heat flow. A vertical scale bar on the right indicates a height of 250 mm. The setup is labeled (b).

(a) : Multi-port Differential Mobility Analyzer (MP-DMA)
(b) : 12th Homemade Condensation Particle Counter (CPC 12)
(c) : M-CPC Module

Figure 1

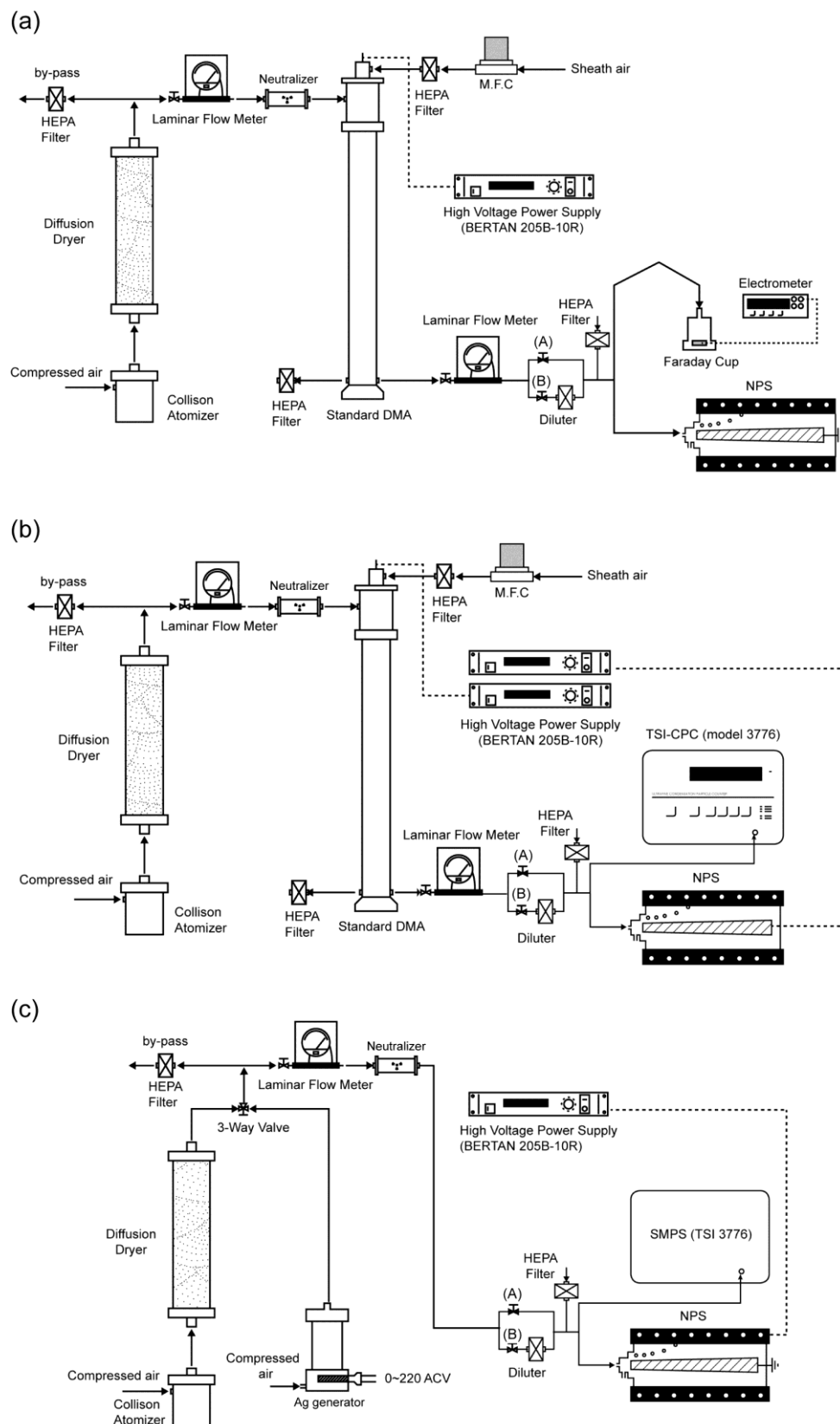
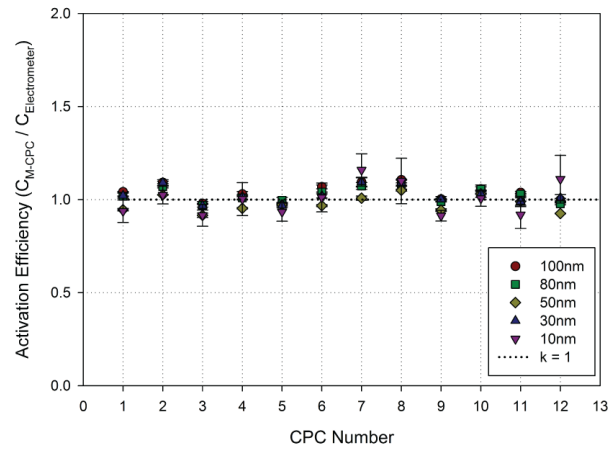


Figure 2

(a)



(b)

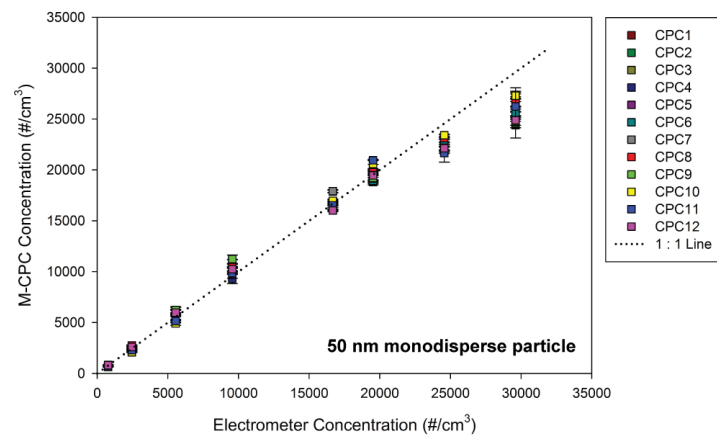
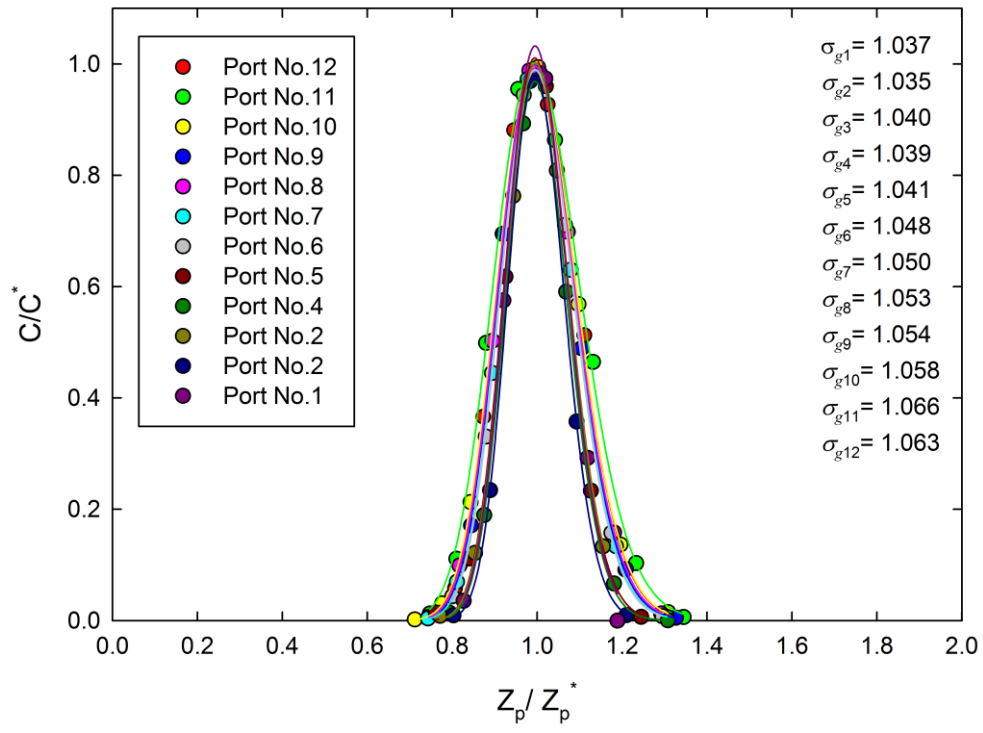


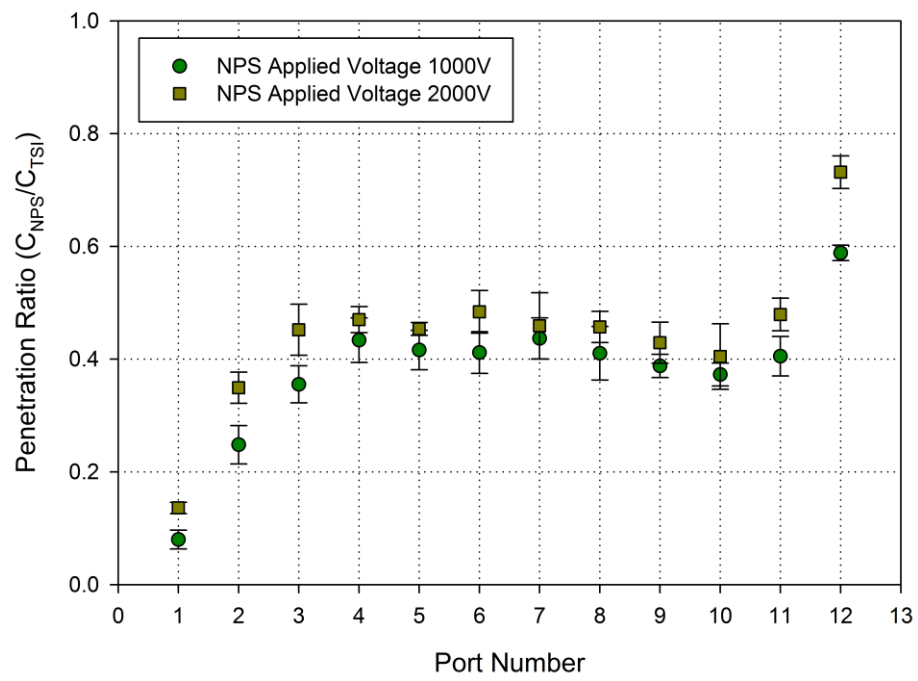
Figure 3



410

411 Figure 4

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414 Figure 5

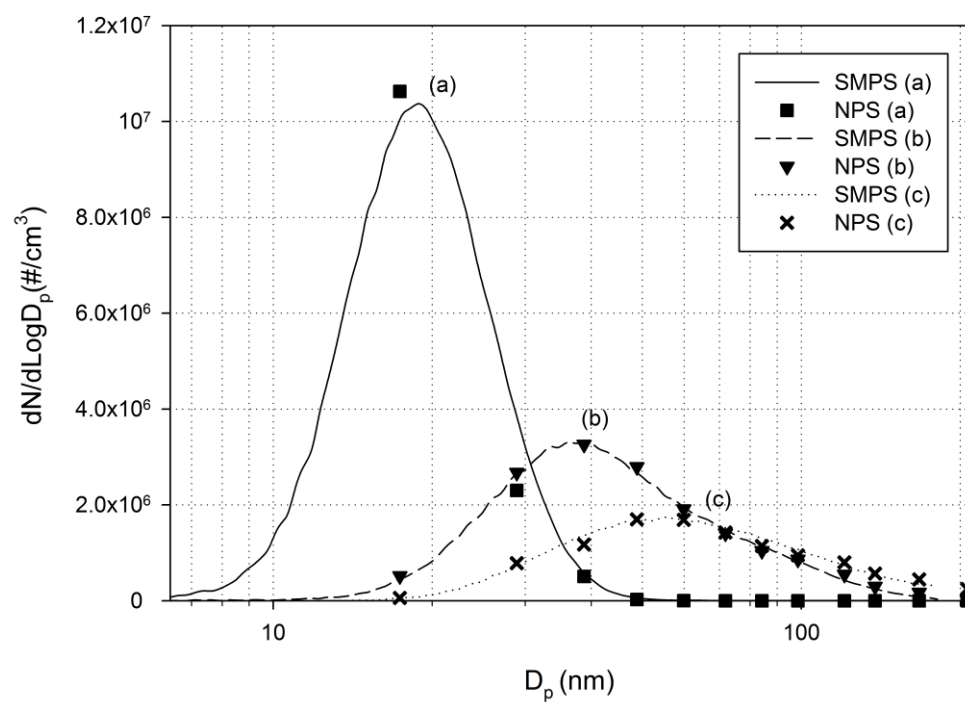
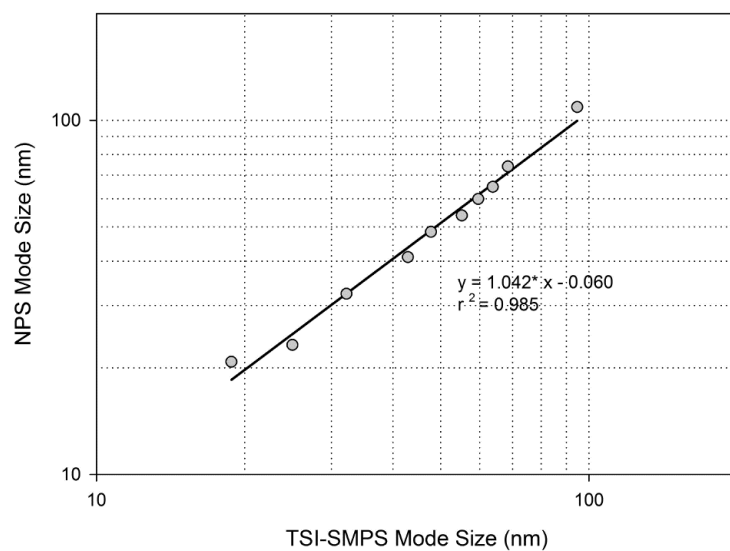
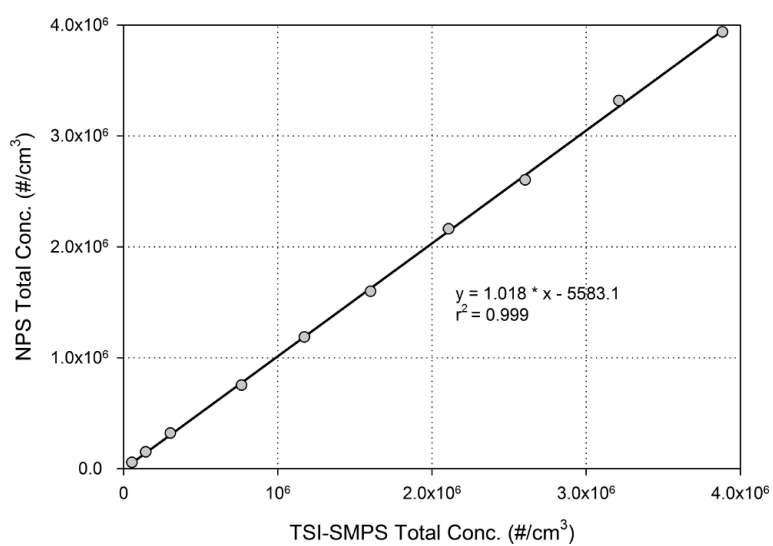


Figure 6

(a)



(b)



418

419 Figure 7

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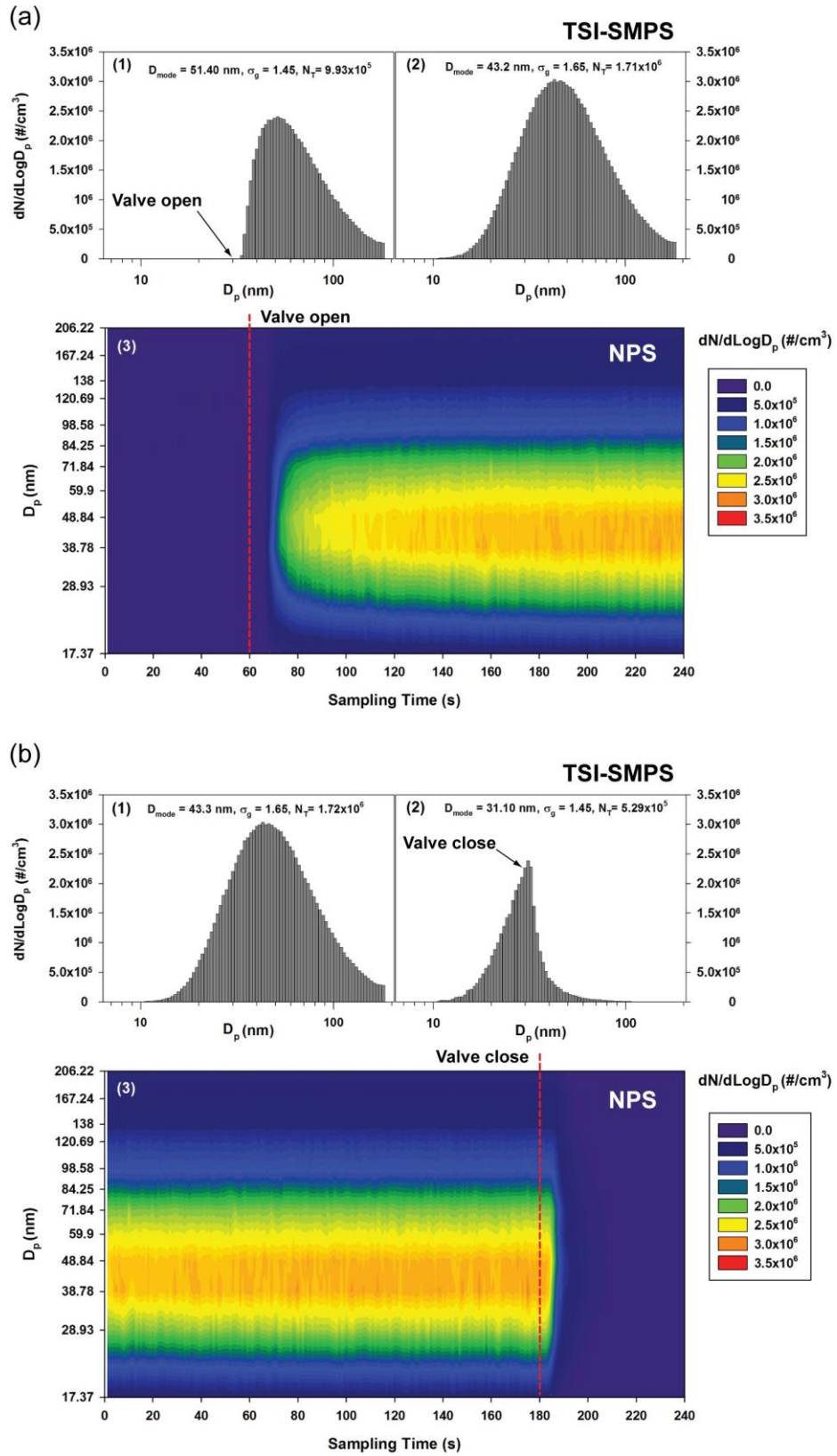


Figure 8

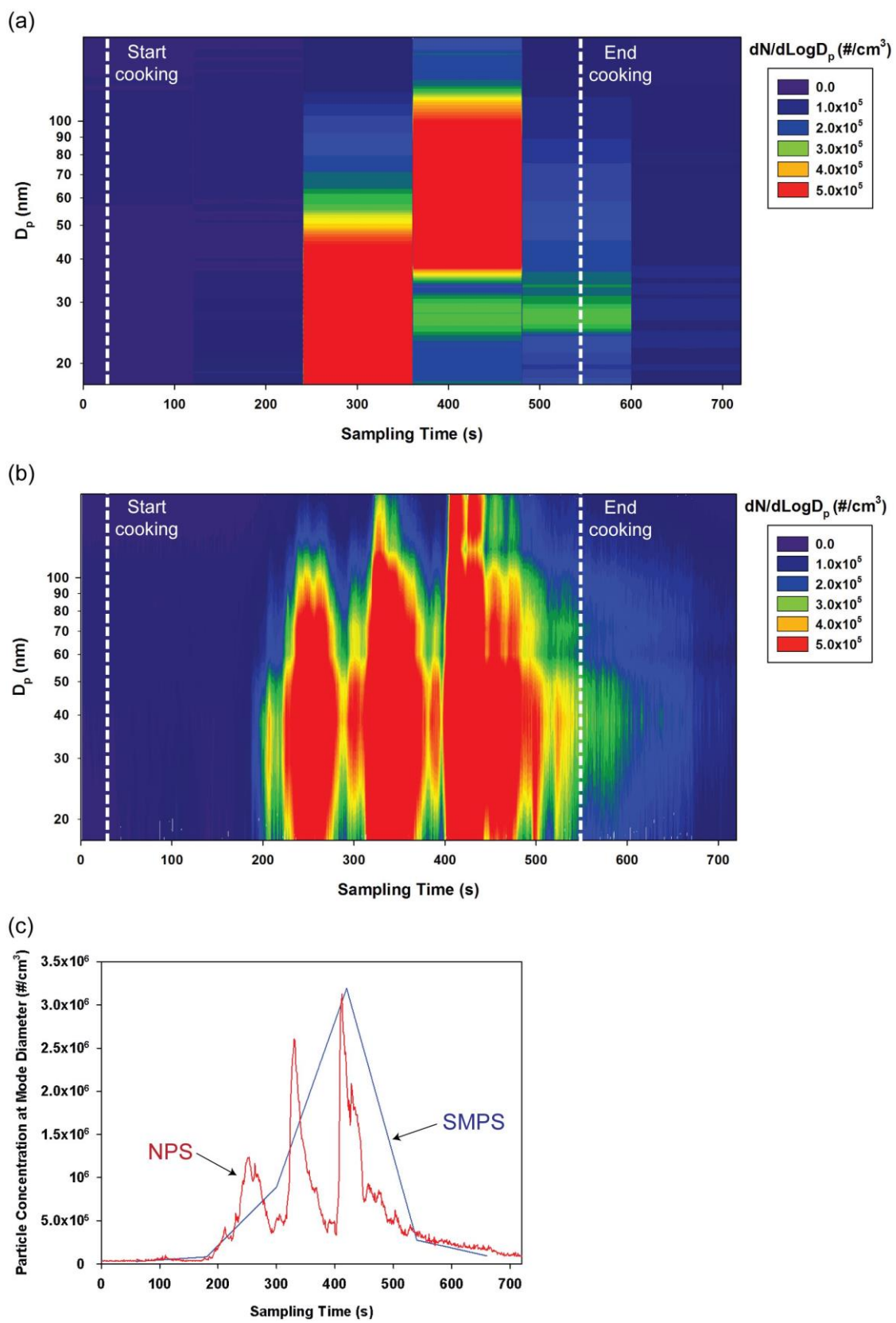


Figure 9

Atmospheric Measurement Techniques (Discussions)

Ref.: Manuscript No. amt-2019-438-RC1

Title: Development of a new Nano-particle sizer equipped with a 12 channel multi-port differential mobility analyzer and multi-condensation particle counters

Anonymous Referee #1, 12 Dec 2019

General comments

The manuscript presents the experimental work on the development of a nano-particle sizer for measuring a particle size distribution in 1 s time resolution. As a fast measurement system for ambient aerosols has attracted attention, this paper has an originality and deals with important contents. In general, the manuscript shall be considered for the journal publication after some major and minor revisions. Specific comments after reviewing the manuscript are given in the following:

Major comments

1. As shown in fig. 1, the inlet of each port seems to be a small hole not an annular ring. Therefore, only a part of introduced particles would be detected by a CPC because particles would be deposited at the wall. So, the particle loss in the MP-DMA might be significant. If particle loss in MP-DMA is high, NPS could not measure low concentration. Then what is minimum measuring concentration of NPS? If the inlet shape of each port is the annular ring, the flow is expected to be deflected. In a typical DMA, the flow deflection is minimized by centering the flow from the annular ring. How did author solve the flow deflection problem in the MP-DMA?

Ans: Thanks for the clarification. In fact, the shape of the sampling ports is annular. We agree that this is not clearly mentioned in the original manuscript. Therefore, we modified the sentence as follows:

Line 94: “While Chen et al. (2007) employed three sampling ports and applied an exponentially increasing distance between neighboring ports to allow a wide size range of particles, the MP-DMA has 12 annular sampling ports that are placed with a uniform distance of 2 cm between neighboring ports.”

2. TSI-SMPS and NPS showed very good agreement for the particle concentration distribution as shown in fig. 7. Can the NPS detect particles smaller than 17 nm by decreasing NPS voltage? Why did not the authors perform the experiment with the voltage lower than 1000 V?

Ans: Thanks for the good comments. We developed the NPS for measuring particle size distribution up to 300 nm particles; therefore, we can utilize the NPS together with an optical particle counter (OPC) for fast ambient particle measurements. As the reviewer mentioned, the NPS might be used at the voltage range under 1000 V to classify smaller particles down to 10 nm. The developed NPS is a prototype, so we are optimizing the flowrate and configuration of the NPS system to characterize smaller particles down to sub-10 nm particles.

3. As shown in fig. 9, while the SMPS immediately responded when an aerosol valve was closed or opened, the NPS has 15-20 s response time. Authors explained it with concentration stabilization and particle

transportation. However, concentration stabilization might not be the reason because the SMPS responded immediately. Furthermore, particle transportation cannot be the reason if the length of transportation pipe of SMPS and NPS were same. It would be only 3 seconds late even considering the flying time in NPS. Why NPS response time was too late?

Ans: Thanks for the good comments. In the data processing, we made a mistake. We did not consider the preparation time (about five seconds) of the SMPS before the scanning process. In the experiments, we clicked the start buttons of SMPS and NPS systems simultaneously. The NPS measures size distribution right after the start; however, the SMPS system takes 4-5 seconds before the scanning process, which was previously not considered in the data processing. Therefore, the NPS measures five seconds prior to the SMPS. Based on this, we corrected Fig. 8 as follows:

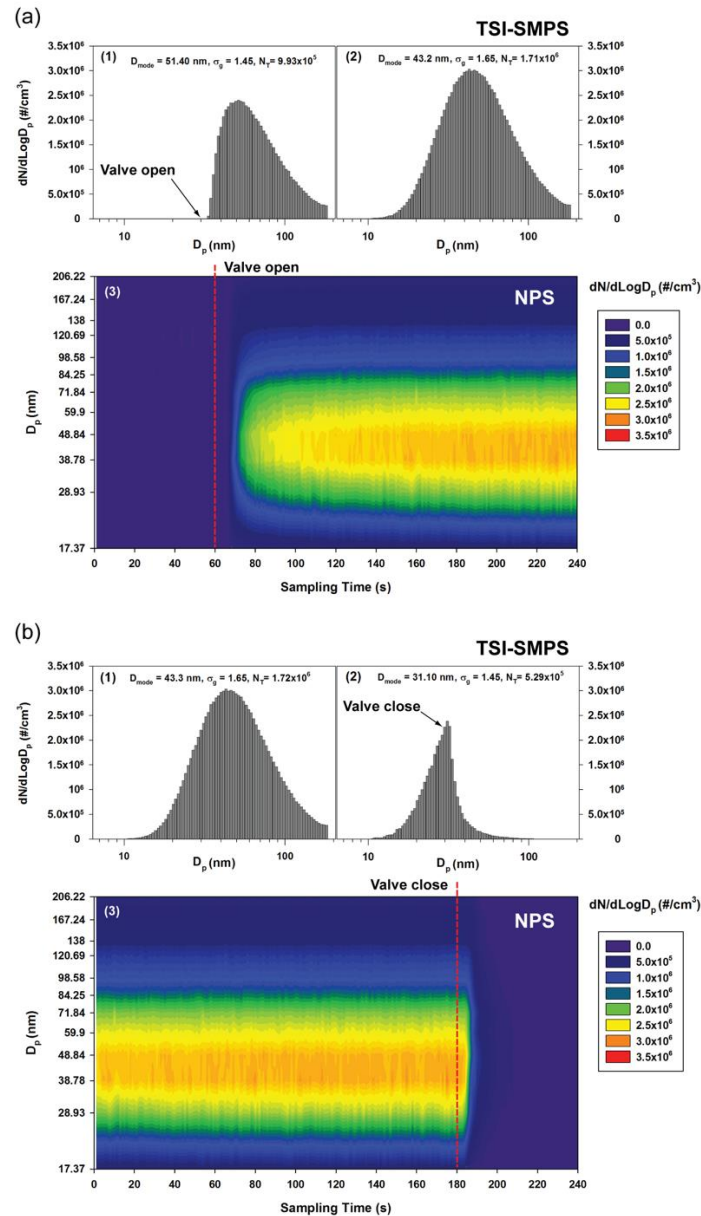


Figure 8. Comparison of the size distributions measured by the TSI-SMPS and NPS for the unsteady particle size distribution in (a) increasing and (b) decreasing particle concentrations. The tested aerosols were introduced or blocked 60 s or 180 s after starting measurements, respectively: (1) the first TSI-SMPS scanning data; (2) the second TSI-SMPS scanning data; (3) the NPS data for 240 s. The data were obtained at the NPS applied voltage of 1000 V.

Furthermore, we provide a graph below on the peak concentration obtained by the NPS as a function of time for two cases (valve open/close). We found that the particle concentration started to increase 5 s after the valve was opened, and the particle concentration started to decrease 5 s after the valve was closed. Notably, the colored scale might not be enough to capture the small changes in concentration, but we confirmed that the NPS takes approximately 5 s to respond to the concentration change. With considering the flight time of approximately 3 s in the classification zone in the NPS as well as the residence time before the flow entering the inlet of the NPS. We found that the observed response time until the signal appears for the TSI-SMPS and NPS seems to be reasonable. Again, thanks for pointing it out, so we could find the mistake in the data processing. We added the plot in the Supplementary Materials.

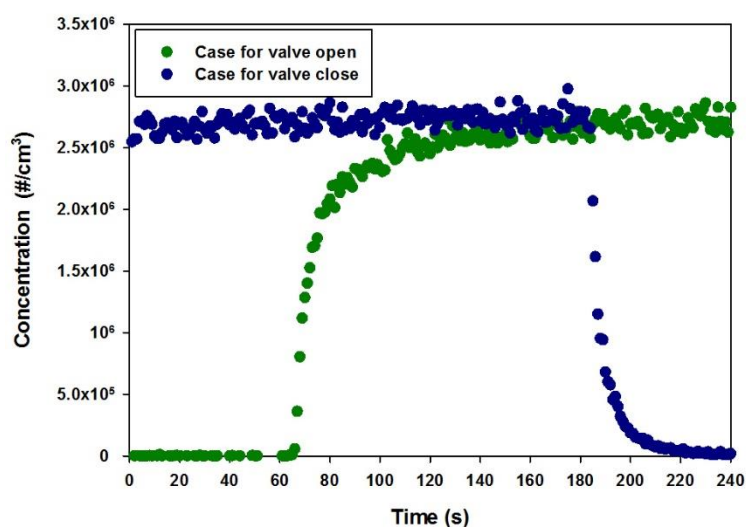


Figure S2. Change in particle concentration at the mode diameter as a function of time.

Line 276: “In Fig. 8(a-1), the concentration data appeared after the valve was opened (60 s after the first scanning process). However, concentrations for particle sizes below 30–32 nm were not shown during the first scanning process because the corresponding lower voltages applied for classifying this size range were already scanned when only the clean air was being measured. In the second scanning process of the TSI-SMPS, the complete size distribution was obtained. The NPS measurement shows that a few seconds after opening the valve, a rapid increase in particle concentration for the complete size range was observed. Specifically, the particle concentration started to increase or decrease approximately 5 s after the valve was opened or closed, respectively (Fig. S2 in Supplementary Material). Considering the response time of the NPS is approximately 3.3 s, (sum of the M-CPC response time of approximately 0.3 s and particle residence time in the MP-DMA, maximum 3 s), the rest of the delay time might be caused by the time required for concentration stabilization and particle transportation. The delay was also observed in the TSI-SMPS. Approximately 2 s after opening the aerosol path (i.e., 1–2 size bins), the concentration started to increase. During the test for rapid decrease in particle concentration (Fig. 8(b)), the performances of the TSI-SMPS and NPS are quite distinct as well. After blocking the particle path 180 s after the measurement, data from the second scanning of the TSI-SMPS show the size distribution for the smaller particles, in a similar manner to the results in Fig. 8(a), because they were already scanned. However, the size distribution measured by the NPS completely disappeared after some delay time. Therefore, the NPS can be successfully used for unsteady particle size distributions to observe changes in concentration.”

Specific comments

1. It seems that the difference between concentrations obtained by the M-CPC and electrometer was insignificant in fig. 3b. However, in fig. 5, the difference between the data from the M-CPC and TSI-CPC is large. Authors should explain why the two cases are different so that the reader will not be confused.

Ans: Thanks for the comments. We obtained the activation efficiency (Figure 3) of the M-CPCs with the zero voltage in the MP-DMA (due to the assembled configuration) in order to examine the performance of the M-CPCs (experimental setup: Figure 2a). Therefore, all the particles introduced to the NPS can be measured by the M-CPCs. The results are shown in Figure 3. We modified some sentences for the better understanding of the part.

Line 192: “To obtain the activation efficiency, monodisperse particles were measured by the electrometer and NPS operated at 0 V as shown in Fig. 2(a). For the NPS measurement, all aerosols were introduced through the sheath flow inlet only (with a flowrate of 3.96 L min^{-1}), so the particle concentrations could be measured by all M-CPCs. The same flowrate of 3.96 L min^{-1} was introduced to the electrometer, and the measurements were carried out simultaneously.”

On the contrary, to obtain the penetration efficiency (Figure 5), we operated the NPS at 1000 V and 2000 V (experimental setup: Figure 2b). We used the penetration efficiency as a correction factor to have the same performance as the TSI-CPC. Each port has a different sizing resolution due to the different aerosol-to-sheath flowrate ratios. Therefore, the penetration ratio of the MP-DMA is increasing with the increasing port number (far from the aerosol inlet). The detail of the description for the penetration ratio can be found as follows:

Line 221: “The penetration ratio of the MP-DMA ranges from 0.099 to 0.765, and these data were used for calibrating the NPS system to convert the raw data obtained by the NPS to the reference concentration data. The theoretical resolution of the MP-DMA decreases from 21 (Port 1) to 10 (Port 12) due to the increasing aerosol-to-sheath flowrate. However, the resolution of the first DMA (TSI standard DMA) is 10 owing to the ratio between aerosol and sheath flowrate of 1:10. Therefore, the CPC at Port 1 might count the particles in the narrower size distribution classified by the first DMA, resulting in a low penetration ratio. Thus, the penetration ratios for all ports were used as correction factors in Eq. (1) to achieve the same concentration as the reference data measured by the TSI-CPC.”

2. It should be good to indicate ‘50 nm monodisperse’ in fig. 3b.

Ans: Thanks for the suggestion. We modified Figure 3 as the reviewer recommended.

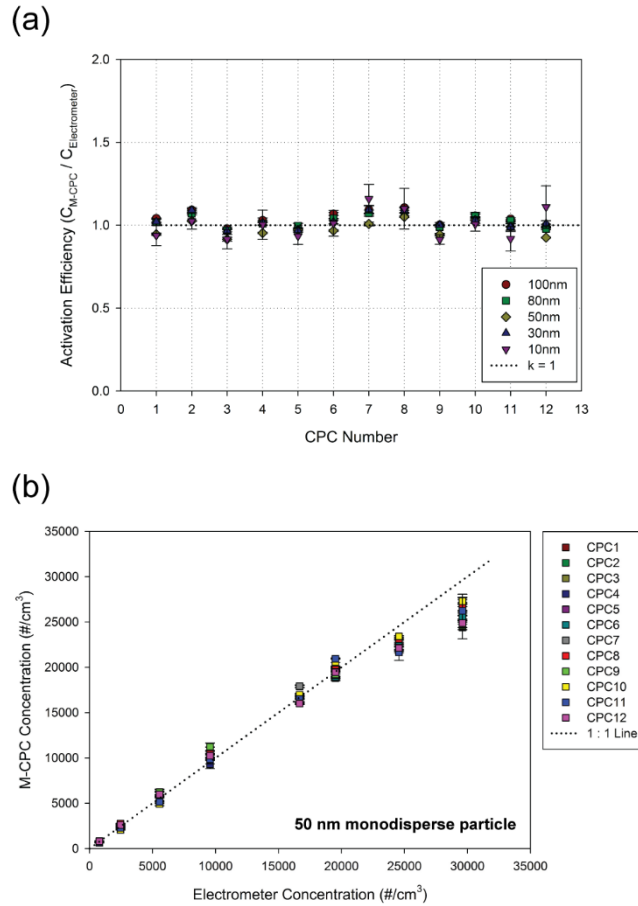


Figure 3. M-CPC performance: (a) activation efficiencies of 12 home-made CPCs; (b) concentration linearity between the electrometer and M-CPCs.

3. It might be better to change fig. 6 to a table.

Ans: Thanks for the recommendation. We removed Figure 6 and changed it to Table 1.

Table 1. Mode diameter of the size distribution obtained by using the central mobility range for each port.

	MP-DMA voltage	Port number											
		1	2	3	4	5	6	7	8	9	10	11	12
Mode diameter [nm]	1000 V	17.4	28.9	38.8	48.8	59.9	71.8	84.3	98.6	120.7	138.0	167.2	206.2
	2000 V	24.4	40.7	54.9	70.1	86.6	103.8	122.9	145.5	177.6	207.9	254.5	315.6

4. It will be better to denote the “valve open” and “valve close” fig. 9 (1) and (2) as well.

Ans: As the reviewer mentioned, we indicated it in Figure 8 (The figure numbering has been changed).

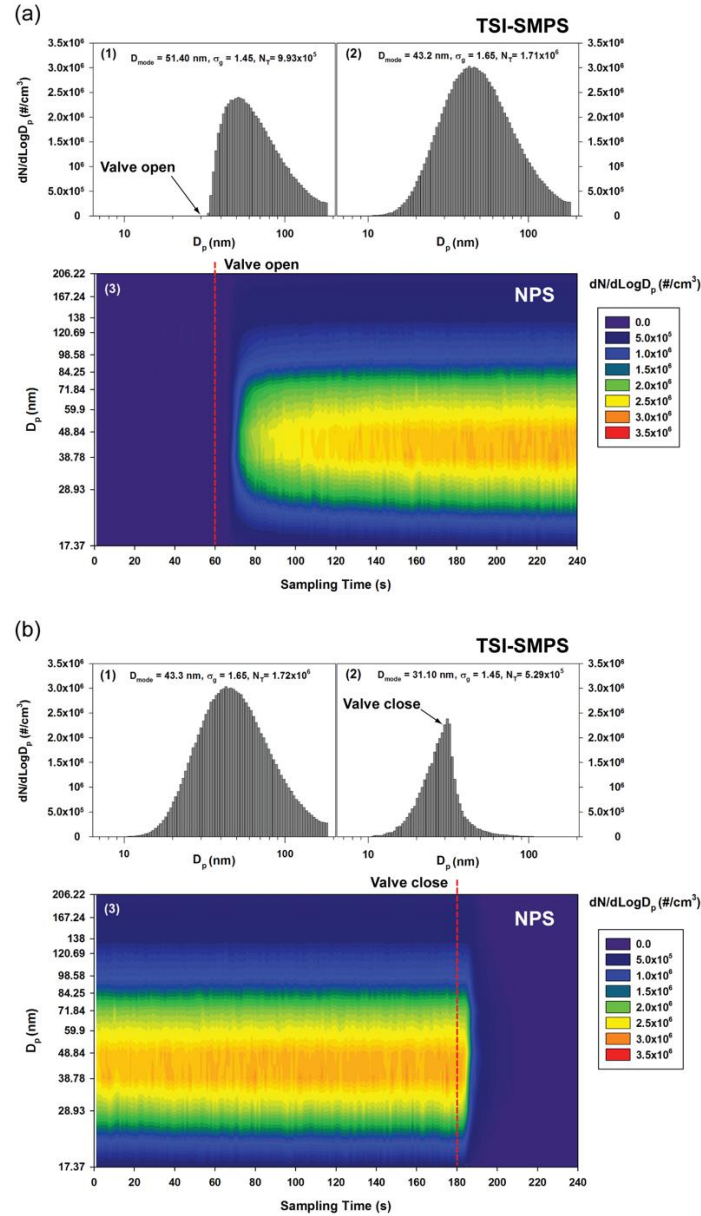


Figure 8. Comparison of the size distributions measured by the TSI-SMPS and NPS for the unsteady particle size distribution in (a) increasing and (b) decreasing particle concentrations. The tested aerosols were introduced or blocked 60 s or 180 s after starting measurements, respectively: (1) the first TSI-SMPS scanning data; (2) the second TSI-SMPS scanning data; (3) the NPS data for 240 s. The data were obtained at the NPS applied voltage of 1000 V.

5. The minor ticks in the x-axis in fig. 9 (1) and (2) are hard to recognize.

Ans: We increased the length of the major and minor ticks in Figure 8(1) and 8(2). Please refer to the answer to the previous question (The figure numbering has been changed).

6. Line 85: The NPS seems to be movable. Then, what is the weight of the NPS? Is it hard to move by human hands or not?

565 Ans: Thanks for the question. The entire system of the NPS is approximately 15 kg. The system can be moved
566 from place to place for sure.

567

568 **7. Line 168: Author mentioned that the maximum flying time of particles inside the NPS is approximately**
569 **3 s. Were the NPS data corrected based on the flying time?**

570 Ans: Thanks for pointing it out. We already considered the delay from the particle residence time. As the reviewer
571 mentioned the larger particles (latter port) take more time to be classified, so when the size distribution is obtained,
572 the delay factor was considered. This is indicated in the revised manuscript.

573 Line 144: “The delay due to the residence time inside the MP-DMA was considered when obtaining the size
574 distributions.”

575

576 **8. Line 182: Z_p is not presented in Eq. (1), but the description of Z_p is shown in the manuscript. Please**
577 **check the equation.**

578 Ans: Sorry for the confusion. We edited the manuscript as follows:

579 Line 186: “where D_p is the particle diameter, f_c is the charge fraction, P is the penetration ratio, and $\eta_{CPC,act}$ and
580 $\eta_{CPC,det}$ are the activation and detection efficiency of the M-CPC, respectively.”

581

582 **9. It might be difficult for the readers to understand and compare contour graphs of the NPS and SMPS in**
583 **fig. 10. It might be better to include in the plot of the obtained mode diameters and concentrations as a**
584 **function of time.**

585 Ans: We added an additional plot for peak particle concentrations as a function of sampling time, as shown in
586 Figure 9(c).

587 Line 300: “Figure 9(c) shows the particle concentration at a peak particle size for each measurement of the TSI-
588 SMPS and NPS.”

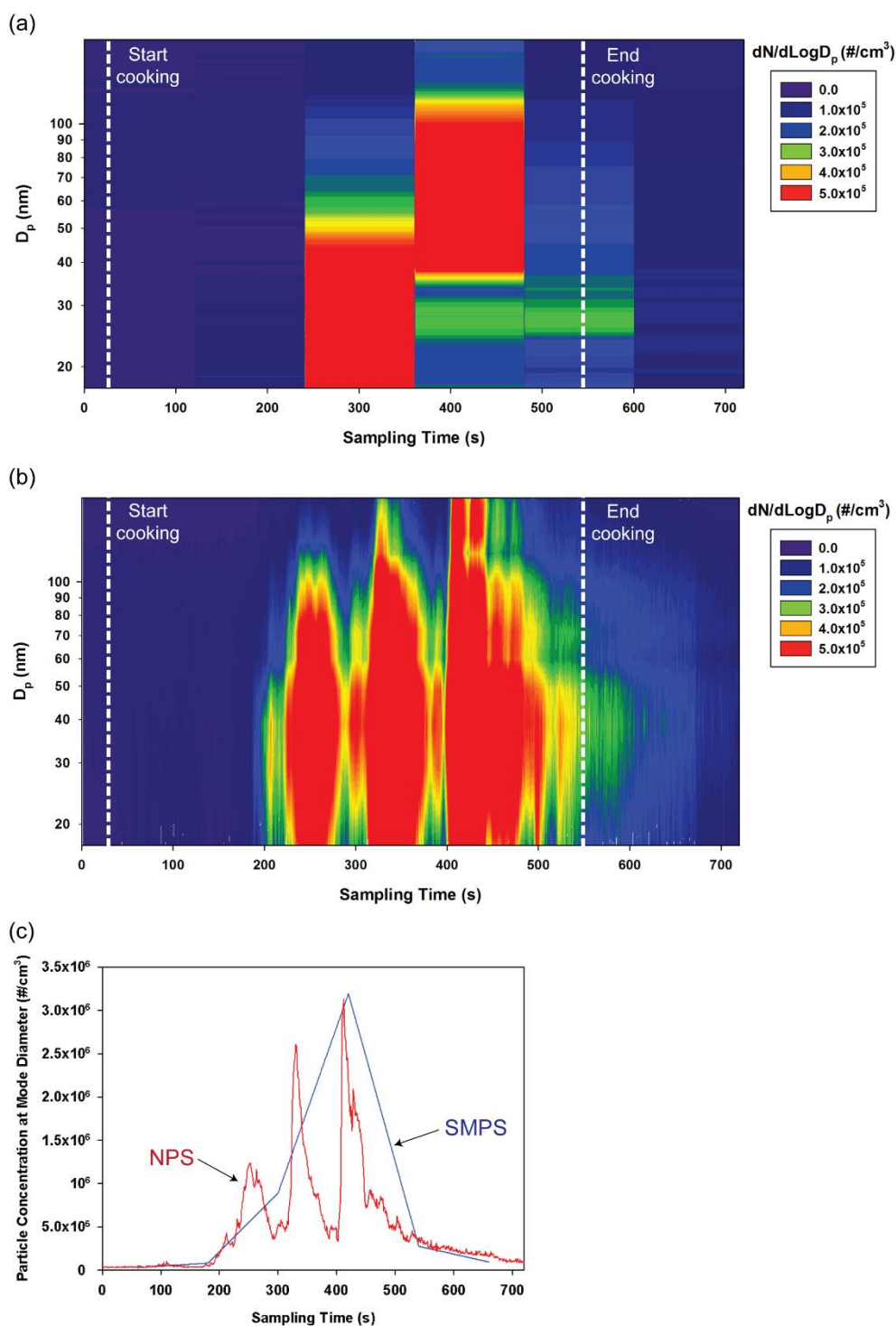


Figure 9. Size distributions measured by the (a) TSI-SMPS and (b) NPS during a cooking activity and (c) variation of particle concentration at mode diameters. The NPS data were obtained at the applied voltage of 1000 V.

10. Line 283: The authors should state the positions of the sampling inlets of the NPS and SMPS. The sampling positions for the two instruments should be close to each other for the reliable data comparison. This should be also mentioned in the manuscript.

Ans: Thanks for the clarification. As the reviewer mentioned the sampling points for the SMPS and NPS measurements are close to each other, approximately 10 to 15 cm. Sampling locations are approximately 1 m away from the cooking spot, and the distance from the ground to the sampling port is around 0.8 m. This sampling location, quite close to the cooking spot (1 m), frequently caused sudden changes in concentration. Based on this information we added the comments on this in the revised manuscript.

Line 296: “The sampling location for the TSI-SMPS and NPS measurements is 1 m away from the cooking spot, which caused sudden changes in concentration.”

11. Line 307: In general, particle concentration of diesel emission or roadside atmospheric particles is high, and the authors mentioned in the conclusion that the NPS can be used in these applications. Furthermore, the authors mentioned that the advantage of the NPS is in measuring low concentration of particles in the introduction when compared to the FMPS. The authors need to clearly state the purpose (or applications) of the NPS.

Ans: Thanks for the good comments. The advantage of the electrical mobility analyzer system (with a condensation particle counter, CPC) such as SMPS and, in this study, NPS is the wide detection range of concentration (low to high concentrations). Therefore, we take an example of vehicle emission studies in terms of fast-changing concentration condition. As the reviewer mentioned, the statement can be confusing. Therefore, we focused more on the fast-changing concentration conditions and changed the sentence as follows:

Line 320: “From the findings in this study, we believe that the NPS can be a promising instrument providing comprehensive information on fast-changing concentration environments.”

General comments

This manuscript presents a design of a novel differential mobility analyzer with multiple outlets enabling fast parallel measurement of particle size. The manuscript is written mostly in a clear and concise manner presenting the main details of the design of the instrument and tests done to verify its operation. However, some parts of the manuscript explaining the experiments need clarification (see questions below). This manuscript is fit for publications once the questions and comments below have been addressed.

1. (Page) 4, L. (Line) 87: Suggest changing wording to help the reader to understand the difference between "aerosol flow rate" and "sampling flow rate". Perhaps "sampling flow rate for each CPC".

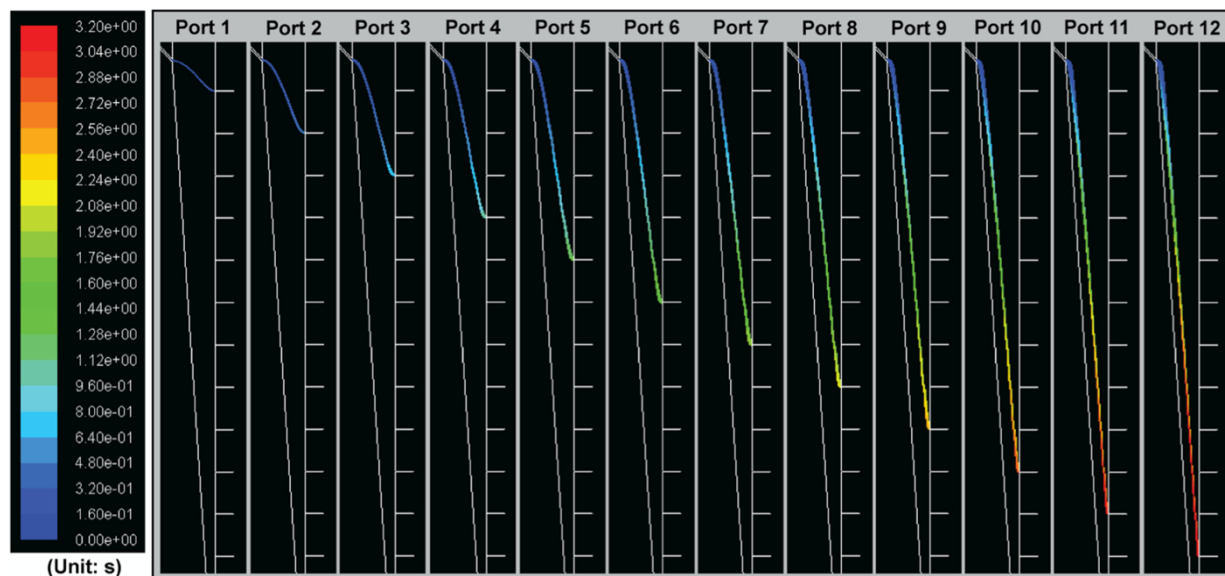
Ans: Thanks for the good suggestion. We modified the sentence as follows:

Line 87: "The flow systems and paths for the NPS are depicted in Fig. 1, including the aerosol flowrate (Q_a , 0.18 L min⁻¹), sheath flowrate (Q_{sh} , 3.78 L min⁻¹), sampling flowrate for each CPC (Q_s , 0.18 L min⁻¹), and exhaust flowrate (Q_e , 1.8 L min⁻¹)."

2. P. 4, L. 95: As each CPC samples through a single port, how uniform are the sample flows across the circumference of each annulus? One would expect needing multiple ports per annulus to ensure uniformity of flows. Was any CFD modelling done to study the internal flows? Please discuss it.

Ans: Thanks for the good comments. The "uniform" in the line 95 (in the original manuscript) means that the annular ports are placed with the uniform distance of 2 cm. We agree that this wording might be confusing to readers. Therefore, we deleted the part. As the reviewer mentioned, we recently performed and published the numerical work on the MP-DMA performance, using the computational fluid dynamics (CFD) tool. The numerical simulation focused on flow field and particle transport inside the MP-DMA. The numerically obtained transmission efficiency and resolution agreed well with experimental data. The figure below represents the particle transport (with particle residence time) obtained after flow field simulation. Furthermore, we expect that the flow through each annulus might be quite uniform owing to the small sampling slit (approximately 0.5 mm), which might result in pressure drop and thus uniform flow through the annular slit. It is not easy to observe and evaluate the uniformity of the flow inside the instrument experimentally, but from the consistent results between the experiments and numerical simulations (transmission efficiency and resolution of the MP-DMA), we can assume that flow inside the NPS should be similar to the flow obtained in the simulation, which does not show any uniformity issue. Based on the reviewer's comment, we put some information in the revised manuscript as follows:

Line 96: "The MP-DMA uses an inner electrode with the increasing diameter along the longitudinal direction."



3. P. 4, M-CPC: are there any publications about the M-CPC which could be referenced in this manuscript? If not, then more information about the design and working parameters of the M-CPC should be provided here.

Ans: Thanks for pointing it out. In our lab, we developed aerosol instruments including condensation particle counter, optical particle counter, differential mobility analyzer, etc. We have been employing our homemade CPC for investigating atmospheric aerosols. It has the same parts including a saturator, condenser, and optical part. We list the references below that employed our CPC.

- (1) Querol, X., Gangoiti, G., Mantilla, E., Alastuey, A., Minguillón, M. C., Amato, F., Reche, C., Viana, M., Moreno, T., Karanasiou, A., Rivas, I., Pérez, N., Ripoll, A., Brines, M., Ealo, M., Pandolfi, M., Lee, H. K., Eun, H. R., Park, Y. H., Escudero, M., Beddows, D., Harrison, R. M., Bertrand, A., Marchand, N., Lyasota, A., Codina, B., Olid, M., Udina, M., Jiménez-Esteve, B., Jiménez-Esteve, B. B., Alonso, L., Millán, M. and Ahn, K. H.: Phenomenology of high-ozone episodes in NE Spain, *Atmos. Chem. Phys.*, 17(4), 2817–2838, doi:10.5194/acp-17-2817-2017, 2017.
- (2) Zhu, Y., Wu, Z., Park, Y., Fan, X., Bai, D., Zong, P., Qin, B., Cai, X. and Ahn, K. H.: Measurements of atmospheric aerosol vertical distribution above North China Plain using hexacopter, *Sci. Total Environ.*, 665, 1095–1102, doi:10.1016/j.scitotenv.2019.02.100, 2019.
- (3) Minguillón, M. C., Brines, M., Pérez, N., Reche, C., Pandolfi, M., Fonseca, A. S., Amato, F., Alastuey, A., Lyasota, A., Codina, B., Lee, H. K., Eun, H. R., Ahn, K. H. and Querol, X.: New particle formation at ground level and in the vertical column over the Barcelona area, *Atmos. Res.*, 164–165, 118–130, doi:10.1016/j.atmosres.2015.05.003, 2015.
- (4) Hwang, I. and Ahn, K. H.: Performance evaluation of conventional type conductive cooling continuous flow compact water-based CPC (Hy-WCPC), *J. Aerosol Sci.*, 113(July), 12–19, doi:10.1016/j.jaerosci.2017.07.007, 2017.

Line 106: “The operating principle of the M-CPC is same as other typical CPCs. Particles are introduced to the saturator (temperature: 35 °C), and the condensational growth of the particles occurs in the condenser at a temperature of 10 °C. The condensed particles are detected in the optical part.”

4. P. 8, L. 210 and below, also start of P. 9: This paragraph needs elaboration with more explanation provided on how the experiment and data analysis was done. For example, what is meant by "central

particle diameter"? How were penetration ratios obtained? Was the TSI SMPS size classification point changed or kept constant? What were the parameters of the aerosol size distribution coming from the SMPS? Was the SMPS data corrected in any way (multiple charging, diffusion losses etc.)? Please add more details.

Ans: Thanks for the comments. The answers are presented below.

4-1) what is meant by "central particle diameter"? How were penetration ratios obtained?

Ans: The central particle size represents the mode diameter of the classified particles at each port as shown in Table 1 below. The penetration ratio is defined as the ratio between the concentrations of monodisperse particles, generated by the DMA in Fig. 2(b), obtained by each M-CPC and TSI-CPC. For example, we generated 17.4 nm monodisperse particles by using a DMA in Fig. 2(b) and measured the concentrations using the NPS operated at 1000 V and TSI-CPC.

Table 1. Mode diameter of the size distribution obtained by using the central mobility range for each port.

	MP-DMA voltage	Port number											
		1	2	3	4	5	6	7	8	9	10	11	12
Mode diameter [nm]	1000 V	17.4	28.9	38.8	48.8	59.9	71.8	84.3	98.6	120.7	138.0	167.2	206.2
	2000 V	24.4	40.7	54.9	70.1	86.6	103.8	122.9	145.5	177.6	207.9	254.5	315.6

Line 217: "The penetration ratio is defined as the ratio of the total concentration at the central particle diameter (ref. Table 1) measured by the NPS to the reference concentration obtained by the TSI-CPC as presented in Fig. 2(b). For example, monodisperse particles with a mode diameter shown in Table 1 were generated by using a DMA and introduced to the NPS and TSI-CPC to achieve the penetration ratio."

Line 221: "The penetration ratio of the MP-DMA ranges from 0.099 to 0.765, and these data were used for calibrating the NPS system to convert the raw data obtained by the NPS to the reference concentration data. The theoretical resolution of the MP-DMA decreases from 21 (Port 1) to 10 (Port 12) due to the increasing aerosol-to-sheath flowrate. However, the resolution of the first DMA (TSI standard DMA) is 10 owing to the ratio between aerosol and sheath flowrate of 1:10. Therefore, the CPC at Port 1 might count the particles in the narrower size distribution classified by the first DMA, resulting in a low penetration ratio. Thus, the penetration ratios for all ports were used as correction factors in Eq. (1) to achieve the same concentration as the reference data measured by the TSI-CPC."

4-2) Was the TSI SMPS size classification point changed or kept constant? What were the parameters of the aerosol size distribution coming from the SMPS? Was the SMPS data corrected in any way (multiple charging, diffusion losses etc.)?

Ans: We employed the TSI-CPC, not TSI-SMPS size classification, for the penetration ratio. As shown in Fig. 2(b) we did not use the scanning-voltage DMA and CPC combination, but we only measured the concentration using the CPC. The DMA in Fig. 2(b) was operated in a fixed voltage mode to generate the monodisperse particles, not scanning voltage mode. The SMPS (scanning voltage mode) was only used for observing the particle size distribution (Fig. 2(c) and from Fig. 7). The SMPS data obtained in this study were corrected based on the multiple charging, charge fraction, and diffusion loss. We used the TSI software to operate the TSI-SMPS system, and this software supports the all the corrections.

Line 261: "For all TSI-SMPS measurements performed in this study, the corrections for the multiple charging and diffusion loss were applied."

5. Figure 2: Is the SMPS in 2(c) the same as "standard DMA" on 2(a) and 2(b)? If so, state it clearly.

Ans: Thanks for the suggestion. Yes, the DMA included in the SMPS system is the same as the DMA used in Fig. 2(a) and 2(b). We clearly denoted it in the revised manuscript. Thanks a lot for the clarification.

Line 248: "The TSI-SMPS system consists of the TSI standard DMA and TSI-CPC which were used in Fig. 2(a) or 2(b)."

6. Figure 3: Is the bias at higher concentrations taken into account in data inversion?

Ans: That is a good question. Thanks. Yes, we included the correction for the bias at higher concentrations based on the results obtained in this study. However, this high concentration range has never been reached in the real applications such as measuring atmospheric particles. The concentration range shown in Fig 3 represents the performance of the M-CPC. From the experiments, we found the detection limit of the M-CPC by introducing the high concentration of aerosols. However, in the real situation, the M-CPC always measures the concentration of particles classified by the MP-DMA. Therefore, the concentration is usually very low because only small fraction of introduced particles (single positively or negatively charged particles) is detected. As the reviewer pointed it out, we put the correction factor in the NPS system in case of the higher concentration introducing to the M-CPC.

Line 204: "It should be noted that a correction factor was considered in the concentration range higher than 20,000 # cm⁻³. Furthermore, each CPC in the NPS always measures the concentration of particles classified by the MP-DMA; therefore, in real applications such as atmospheric particle measurements, this high concentration after classified by the MP-DMA can be rarely achieved."

7. P. 9, L. 248 and Figure 8: There's a 5500 cm⁻³ bias between the total number concentration measurements from the two instruments, with NPS measuring lower than SMPS. Where does this difference originate from? Is this corrected in data analysis/inversion? Does this mean that the NPS can't measure total particle number concentrations less than 5500 cm⁻³? That's a fairly high number for many atmospheric applications. Please discuss.

Ans: As the reviewer mentioned, we observed a 5500 # cm⁻³ bias for the NPS measurement compared to the TSI-SMPS total concentration. We believe that the difference originates from the loss inside the NPS. Due to the low sampling flowrate of 0.18 L min⁻¹ for each CPC, there might be additional diffusion loss. We are now optimizing the flowrate and trying to minimize the loss inside the system by increasing the flowrate control system. Therefore, in the future we believe that the bias will be reduced. Thanks a lot for the good comments again. Based on the reviewer's comment, we added a sentence in the revised manuscript.

Line 257: "As shown in Fig. 7(b), we observed the approximately 5500 # cm⁻³ bias in the total concentration for the NPS measurement compared to the TSI-SMPS. We believe that this originates from the particle loss inside the NPS due to the low sampling flowrate for each CPC in the NPS system."

8. Figures 7, 8, 9: Were any corrections applied to the SMPS data (multiple charging, diffusion losses etc.)? State this clearly to help the reader make accurate assessments of the results.

Ans: We used the TSI-SMPS system by using the package and the software that the TSI company provides. Therefore, in the system there are options for the multiple charging and diffusion loss corrections. We turned on the corrections when we obtained the data in this study. We stated this in the revised manuscript. Thanks for the good suggestion.

Line 261: “For all TSI-SMPS measurements performed in this study, the corrections for the multiple charging and diffusion loss were applied.”

9. Figure 9: What is meant by first and second scanning data in the figure caption? If these are SMPS scans taken during the measurement, then indicate when they were taken on the NPS color plot. Also, please label the individual plots clearly to indicate from which instrument they are from.

Ans: Thanks for the comments. In the experiments, we compared the NPS and TSI-SMPS measurements. Two cycles of the TSI-SMPS measurement were performed consecutively with 120 s scanning time for each cycle, and the NPS obtained concentration data every 1 s. Therefore, “first” and “second” in the figure caption represent the first cycle of 120 s and the second cycle of 120 s for the TSI-SMPS measurements, respectively. The left figure in Fig. 8(a) (we changed the numbering from Fig. 9 to Fig. 8 in the revision process) represents the TSI-SMPS data from the first cycle, and the right figure in Fig. 8(b) shows the data from the second cycle. As the reviewer mentioned, we modified the figure, so it can be more clear to readers.

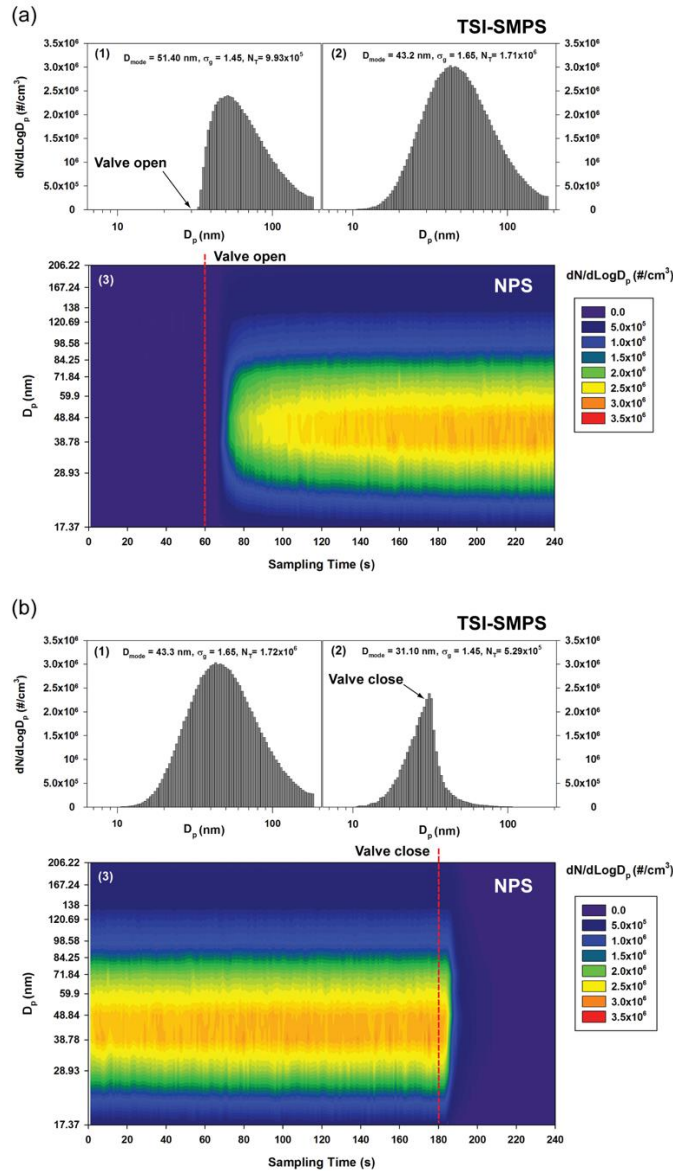


Fig. 8

776 **Response to Editor's comments**

777 Line 10, change "distribution" to "distributions"

778 Done.

779 Line 11, change "distribution" to "distributions"

780 Done.

781 Line 11, change "concentration" to "concentrations"

782 Done.

783 Line 17, I'm not sure this sentence about standard deviations is needed in the abstract. Maybe a statement about
784 the size resolution would be more pertinent.

785 We agree that the comment on the standard deviations is not necessary, so we deleted the part.

786 Line 23, change "For the last" to "Finally, we present NPS measurement results. . . "

787 Done.

788 Line 54, change "particle trajectories" to "droplets nucleated from these spatially separated particles.

789 Done.

790 Line 54, this implies that the FIMS detects sub-10 nm particles; it does not.

791 Thanks for the comment. We modified the sentence to "The FIMS can be used to obtain size distributions at sub-
792 second time intervals."

793 Line 85, change "sampling ports" to "sampling ports (annular slits)".

794 Done.

795 Line 87, change "with the increasing diameter" to "with increasing diameter"

796 Done.

797 Line 109, remove "In this article".

798 Done.

799 Line 122, can you really determine the mode diameter to within 0.01 of a nm?

800 No, it was obtained from the regression line, so we changed the sentence to "The mode size and geometric standard
801 deviation of the atomized aerosols were 43 nm and 1.65, respectively."

802 Line 215, add "with increasing port number" to the end of the sentence.

803 Done.

804 Line 232, are you making a rectangular approximation of full-width at half-max (FWHM), and thus need to apply
805 the factor of 2?

806 The transfer function is assumed to be triangular for the MP-DMA, so we applied the factor of 2 for obtaining
807 concentrations of entire particles entering sampling slits.

808 Line 257. I doubt that you actually have a bias of this magnitude--at zero particles you don't count $5500/\text{cm}^3$, do
809 you? The uncertainty on the intercept must be large enough to explain this. Suggest removing this whole
810 discussion of intercept bias, or confirming that the uncertainty in the fit of the intercept encompasses zero.

811 Yes, you are right. The part might confuse the readers, so we deleted the discussion on the bias. Thanks for the
812 comment.

813 Line 267, change to "particle size distributions by employing an on/off valve ('A' in Fig. 2(c)) to introduce. . . ."

814 Done.

815 Line 276, change to "60s after the first SMPS scan began."

816 Done.

817 Lines 277-278, change to "for particle sizes <32 nm were not recovered from the inversion of this scan because
818 the corresponding voltages were applied to the DMA before the valve was opened, when there were no particles
819 in the sample line."

820 Done.

821 Line 279, change "scanning process of the TSI-SMPS" to "scan".

822 Done.

823 Line 280, change to "In contrast, the NPS measurement shows a rapid increase in particle concentration for the
824 complete size range soon after the valve was opened."

825 Done.

826 Line 285, change "transportation" to "transport". Also suggest removing the entire following 2 sentences,
827 beginning with "The delay was" and ending with "started to increase."

828 Done.

829 Line 288, change to "TSI-SMPS and NPS were quite distinct as well. After closing the aerosol valve ~ 180 s
830 after"

831 Done.

832 Line 289 change to "from the second scan of the TSI-SMPS showed only smaller particles, in a manner similar to
833 the results in Fig. 8(a)."

834 Done.

835 Line 290, change to "completely disappeared after some delay time. Therefore," to "quickly approached zero.
836 These tests indicate that the NPS can be. . . ."

837 Done.

838 Line 294. Place "Further" before "NPS measurements"

839 Done.

840 Line 295, change to "TSI-SMPS and NPS, respectively, during the cooking of fish."

841 Done.

842 [Line 296. Change "is" to "was"](#)
843 Done.

844 [Line 300. Change to, "SMPS analysis provides only discontinuous size distributions."](#)
845 Done.

846 [Line 308. Remove "In this research"](#)
847 Done.

848 [Line 313 Change "distribution" to "distributions"](#)
849 Done.

850 [Line 314. Change "the" to "a".](#)
851 Done.

852 [Line 316. Remove "the" before "steady-state" and before "unsteady".](#)
853 Done.

854 [Line 317. Add an "s" to "distribution" and "concentration"](#)
855 Done.

856 [Line 321. Change "can be" to "is"](#)
857 Done.

858 [Line 321. Add "for" between "instrument" and "providing"](#)
859 Done.

860 [Line 322. Change to "information on particle size distributions in fast-changing concentration environments."](#)
861 Done.

862