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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

9 Measuring particle size distributions precisely is an important concern in addressing environmental and human 10 health-related issues. To measure particle size distribution, a scanning mobility particle sizer (SMPS) is often used. 11 However, it is difficult to analyze particle size distribution under fast-changing concentration conditions because 12 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 13 14 mobility analyzer (MP-DMA) with 12 sampling ports and multi-condensation particle counters (M-CPCs) that 15 simultaneously measure concentrations of particles classified by the sampling ports. The M-CPC can completely 16 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. For particle classification tests on the 17 MP-DMA, geometric standard deviations of the size distributions of classified particles were estimated in the 18 19 range of 1.035-1.066. We conducted size distribution measurements under steady-state conditions using an 20 aerosol generator and under unsteady conditions by switching the aerosol supply on/off. The data obtained by the 21 NPS corresponded closely with the SMPS measurement data for the steady-state particle concentration case. In 22 addition, the NPS could successfully capture the changes in particle size distribution under fast-changing particle 23 concentration conditions. For the last, we presented the NPS measurement results of size distributions in common 24 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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32 1 Introduction

33 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 34 measurement procedure of this technique begins with a voltage applied to the DMA to classify monodisperse 35 particles in a narrow electrical mobility range, and then the CPC measures the particle number concentration 36 37 (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 38 39 process are required for accurate estimation of the complete size distribution, making the DMPS unable to respond 40 accurately if the concentration is changing rapidly. For this reason, the DMPS method has limited applications. 41 Wang and Flagan (1990) developed a scanning mobility particle sizer (SMPS) to reduce the measurement time. 42 For the SMPS measurement, the applied voltage is increased (or decreased) continuously, and particles 43 consecutively classified by a DMA are counted by a CPC. As a result, measurement time can be reduced to less 44 than two minutes. However, it is still too long to analyze fast-changing particle size distributions. Recently, several 45 aerosol instrument systems have been developed and studied with the aim of faster measurement. A fast mobility 46 particle sizer (FMPS) was developed based on a principle similar to the SMPS system, the electrical mobility 47 analyzer. Instead of a CPC, the FMPS uses multiple electrometers for particle detection, and the system provides 48 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 # cm^{-3}). In 49 addition, current leakage and electrical noise of electrometers sometimes result in less precise measurements. A 50 51 new fast integrated mobility spectrometer (FIMS) for real-time measurement of aerosols was developed (Kulkarni 52 and Wang, 2006). The FIMS detects charged particles based on their different electrical mobilities, which result 53 in different trajectories. A fast charge-coupled device (CCD) imaging system is employed to capture the locations 54 of particle trajectories. The FIMS is capable of excellent activation efficiencies for sub-10 nm particles and can 55 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 56 57 for particle size distribution measurement with high-time resolution. Therefore, it can be used to observe very fast 58 aerosol growth, especially in the sub-10 nm range. However, the DMA-train contains six commercial CPCs and 59 six commercial DMAs, which make the system costly and bulky. Recently, Oberreit et al. (2014) performed 60 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.

68 In addition to the above-mentioned instruments, Chen et al. (2007) and Kim et al. (2007) previously developed 69 a differential mobility analyzer with multiple sampling ports for a fast measurement system. However, the multi-70 stage DMA (MDMA) by Chen et al. (2007) has only three sampling ports and needs three CPCs. Furthermore, an 71 exponentially extended longitudinal length is required to increase the number of sampling ports and accommodate 72 the wide size range of particles. As a result, the system becomes complicated and expensive. Kim et al. (2007) 73 developed a DMA with a multi-port system, a substitution for the MDMA system, and it can classify a total of 74 seven sizes simultaneously. They evaluated the DMA system using monodisperse particles and deduced from the 75 experiments that increasing the number of sampling ports did not affect the classification efficiency and transfer 76 functions of the DMA. This was also theoretically supported in research by Giamarelou et al. (2012), in deriving 77 analytical expressions for estimating the transfer functions and the resolutions of DMAs with multiple sampling 78 ports. However, there is still a lack of research on a fast measurement system that retains the traditional DMA 79 function. Therefore, in this study, we developed a new Nano-particle sizer (NPS), consisting of a multi-port DMA 80 (MP-DMA) and multi-CPCs (M-CPCs), that can perform fast measurement of particle size distributions.

81 2 Instrument

82 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 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.

93 2.2 Design Concept of the MP-DMA

94 While Chen et al. (2007) employed three sampling ports and applied an exponentially increasing distance 95 between neighboring ports to allow a wide size range of particles, the MP-DMA has 12 annular sampling ports 96 that are placed with a uniform distance of 2 cm between neighboring ports. The MP-DMA uses an inner electrode 97 with the increasing diameter along the longitudinal direction. As the diameter of the electrode increases, the 98 distance between the inner electrode and the outer cylindrical electrode decreases. Accordingly, the electrical field 99 strength applied to particles increases as they flow to the downstream side. As a result, the MP-DMA can 100 accommodate a wider size range of particles without excessive extension of the electrode length found in the 101 common cylindrical electrode.

102 **2.3 Design Concept of the M-CPC**

103 Each sampling port in the MP-DMA is directly connected to the inlet of each homemade CPC. Classified particles 104 are introduced to and measured by the CPC. One M-CPC module consists of six homemade CPCs, and the NPS 105 has two M-CPC modules (12 CPCs). The module has a unified saturator and condenser block to maintain uniform 106 temperatures. A common working fluid reservoir is located beneath the saturator block. The operating principle 107 of the M-CPC is same as other typical CPCs. Particles are introduced to the saturator (temperature: 35 °C), and 108 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. In this article, each homemade CPC was denoted as CPC1, CPC2, CPC3, 109 etc., based on their location. CPC1 is closest to the aerosol inlet and CPC12 is closest to the sheath outlet in the 110 111 MP-DMA. The reference CPC used in this study is denoted as TSI-CPC (model 3776, TSI Inc., Shoreview MN, 112 USA).

113

Figure 1

- 114 **3** Experimental Setup and Operating Conditions
- 115 **3.1 M-CPC**

116 In order to evaluate the performance of the M-CPC, the activation efficiency and concentration linearity of each 117 homemade CPC were obtained from comparison with a reference electrometer. Figure 2(a) is the schematic 118 diagram of the M-CPC performance test. Using a homemade Collison atomizer, a 0.1 wt% NaCl solution was 119 atomized, and the aerosols were classified by the first DMA (standard DMA, model 3081, TSI Inc., Shoreview 120 MN, USA) to generate monodisperse particles which were distributed to the analyzing instruments. In this study, 121 the operating sheath and aerosol flowrates in the first DMA were 10 L min⁻¹ and 1 L min⁻¹, respectively. The mode 122 size and geometric standard deviation of the atomized aerosols were 43.22 nm and 1.65, respectively. The particle 123 sizes obtained from the atomizer were smaller than 100 nm, thereby minimizing multiple charging effects on the 124 size-selection (Fig. S1 in Supplementary Material). The concentration of particles was controlled by a diluter 125 before entering the instruments as shown in Fig. 2(a). To measure the particle number concentration as a reference, 126 an electrometer (model 6517A, Keithley) with a Faraday cup was used. This is one of the most commonly used 127 methods for CPC calibration (Liu and Pui, 1974). In this experiment, the sampling flowrate of each CPC was 0.18 128 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 129 130 number concentration every 1 s, and the response time of the M-CPC is less than 0.3 s. The experimental setup 131 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 132 133 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 134 135 measured by each CPC and the electrometer were compared.

136 **3.2 MP-DMA**

137 To evaluate the performance of the MP-DMA, the normalized particle mobility distribution for each port and 138 penetration efficiency for the MP-DMA were obtained. Figure 2(b) is the schematic diagram of the MP-DMA 139 performance test. The particle size and concentration were controlled by the first DMA and dilutor, respectively. 140 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 141 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 142 the downstream side because each CPC takes 0.18 L min⁻¹. Under these flow conditions, the residence time of the 143 particles flowing from the aerosol inlet to each sampling port inlet is approximately 0.3 s (Port 1) to 3 s (Port 12) 144 (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 # cm⁻³ by adjusting the valve ('B' in Fig. 2(b)) located in the diluter.

150 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 151 152 concentrations were normalized by the maximum concentration for each port. The electrical mobility was 153 normalized by the central mobility for each port, and the results are shown in Fig. 4. In addition, the particle 154 penetration ratios as a function of port number at voltages of 1000 V and 2000 V are shown in Fig. 5, representing 155 the maximum ratio between the measured concentration at each CPC and the upstream concentration measured 156 by the TSI-CPC, which is approximately $10,000 \ \# \ cm^{-3}$. The maximum penetration ratio was obtained at the 157 central electrical mobility for each port. The penetration ratios were used to calibrate the NPS data in the inversion 158 process.

159 **3.3 Particle Size Distribution Measurement**

160 To test the performance of the NPS, the experimental set-up in Fig. 2(c) was used. For particle generation, we 161 used two types of particles, NaCl and Ag. The NaCl and Ag particles were generated by the homemade Collison atomizer and evaporation generator (Hwang and Ahn, 2017). The particles were neutralized by a neutralizer, and 162 163 the concentration was controlled by a dilutor. The particles were introduced into the TSI-SMPS and NPS. The 164 TSI-SMPS consists of the standard long DMA (model 3081, TSI Inc., Shoreview MN, USA) and a CPC (model 165 3775, TSI Inc., Shoreview MN, USA), and the voltage was generated by a high-voltage power supply 166 (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 167 with constant NaCl and Ag particle concentrations and with changing NaCl particle concentrations during the 168 transition to the equilibrium state. To provide unsteady particle concentrations, we used the on/off valve at the 169 170 aerosol path ('A' in Fig. 2(c)) before the TSI-SMPS and NPS. The total measurement time was 240 s. Two cycles 171 of the TSI-SMPS measurement were performed consecutively with 120 s scanning time for each cycle, and the 172 NPS obtained concentration data every 1 s.

Figure 2

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176 **3.4 Inversion Process for the NPS Concentration Data**

177 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 178 179 through the MP-DMA. The real concentration of each sampling port was estimated by Eq. (1), and the multiple 180 charge correction was referred by Hoppel's inversion method (Hoppel, 1978). Variables used in this inversion 181 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 182 distribution (Wiedensohler, 1988). For a clear understanding of the variables in Eq. (1), we added a brief 183 184 explanation of the experimental method in each result section.

185
$$\left. \frac{dN}{d\log D_p} \right|_{D_p^*} = \frac{2 \times N_{raw}(D_p^*)_n \times (60/1000)}{f_C(D_p^*)_n \times P(D_p^*)_n \times \eta_{CPC,dct}(N_{paw})_n \times \left\{ \log(D_{p,E})_n - \log(D_{p,S})_n \right\}}$$
(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.

190 4 Result and Discussion

191 **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.

200 We also examined the detectable concentration range for the M-CPCs using the experimental setup in Fig. 2(a). 201 The test was conducted with 50 nm monodisperse particles under different concentration conditions. The 202 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 # cm⁻³, indicating that each homemade CPC can 203 be used for concentrations up to this value. It should be noted that a correction factor was considered in the 204 205 concentration range higher than 20,000 # cm⁻³. Furthermore, each CPC in the NPS always measures the 206 concentration of particles classified by the MP-DMA; therefore, in real applications such as atmospheric particle 207 measurements, this high concentration after classified by the MP-DMA can be rarely achieved.

208

Figure 3

209 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.

216

Figure 4

217 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) 218 219 measured by the NPS to the reference concentration obtained by the TSI-CPC as presented in Fig. 2(b). For 220 example, monodisperse particles with a mode diameter shown in Table 1 were generated by using a DMA and 221 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 222 the NPS to the reference concentration data. The theoretical resolution of the MP-DMA decreases from 21 (Port 223 224 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 225 1 might count the particles in the narrower size distribution classified by the first DMA, resulting in a low 226 penetration ratio. Thus, the penetration ratios for all ports were used as correction factors in Eq. (1) to achieve the 227 228 same concentration as the reference data measured by the TSI-CPC. Notably, in this experiment, the reference 229 data are the concentrations of particles classified by the first DMA, and thus the shape of the input particle size 230 distribution is close to a triangle. Therefore, N_{raw}/P (measured raw concentration divided by the penetration ratio) 231 represents the area under a triangle. For this reason we multiplied a factor of 2 as shown in Eq. (1) assuming that 232 a shape of the size distribution of particles entering each port in the NPS is rectangular.

233

Figure 5

234 Table 1 summarizes the central particle diameters on each port under different voltage conditions, 1000 V and 235 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 236 237 and 25–320 nm at 2000 V. The range can be easily adjusted by changing the applied voltage of the NPS. However, 238 there still remains a limitation in the MP-DMA. There is a blank area between Port 1 and Port 2 where particles 239 with a geometric standard deviation less than 1.04 (narrow size distribution) and a mode diameter between those 240 of Port 1 and Port 2 are deposited and will not be detected. However, most real-world aerosol systems have a wide 241 range of size distribution. Furthermore, the size distribution of aerosols with a geometric standard deviation of 242 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. 243

244

Table 1

245 **4.3 Performance of the NPS**

246 **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 252 generator. The data points from the NPS measurements agree with the TSI-SMPS data. Because the NPS has 12 253 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 254 255 of the fitting method used in this study, we also measured different sets of polydisperse particles (total of ten size 256 distributions) using the TSI-SMPS and NPS to obtain the mode size and total concentration of each size 257 distribution, represented in Fig. 7(a) and 7(b). As shown in Fig. 7(b), we observed the approximately 5500 # cm⁻ 258 ³ bias in the total concentration for the NPS measurement compared to the TSI-SMPS. We believe that this 259 originates from the particle loss inside the NPS due to the low sampling flowrate for each CPC in the NPS system. 260 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 261 262 for the multiple charging and diffusion loss were applied.

263

Figure 6

264

Figure 7

265 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 266 267 unsteady particle size distribution by employing the on/off valve to introduce or block aerosols to the instruments. 268 Fig. 8 shows the comparison between the particle size distributions obtained by the TSI-SMPS and NPS for 240-269 s measurements. The dotted red line in Fig. 8(a-3) represents the moment we opened the valve ('A' in Fig. 2(c)), 270 indicating introduction of aerosols 60 s after the beginning of the first TSI-SMPS (or NPS) measurement. In Fig. 271 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 272 273 diameter and number concentration, respectively. The NPS data is represented in a contour graph with the 274 sampling time (x-axis) and particle diameter (y-axis). The color indicates the particle number concentration measured by the NPS. 275

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

280 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 281 or decrease approximately 5 s after the valve was opened or closed, respectively (Fig. S2 in Supplementary 282 283 Material). Considering the response time of the NPS is approximately 3.3 s, (sum of the M-CPC response time of 284 approximately 0.3 s and particle residence time in the MP-DMA, maximum 3 s), the rest of the delay time might 285 be caused by the time required for concentration stabilization and particle transportation. The delay was also 286 observed in the TSI-SMPS. Approximately 2 s after opening the aerosol path (i.e., 1–2 size bins), the concentration 287 started to increase. During the test for rapid decrease in particle concentration (Fig. 8(b)), the performances of the 288 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 289 290 to the results in Fig. 8(a), because they were already scanned. However, the size distribution measured by the NPS 291 completely disappeared after some delay time. Therefore, the NPS can be successfully used for unsteady particle 292 size distributions to observe changes in concentration.

293

Figure 8

294 NPS measurements under unsteady conditions of rapid-changing particle concentrations were performed for real-295 world applications. Figure 9(a) and 9(b) represent particle size distributions measured by the TSI-SMPS and NPS 296 during cooking fish, respectively. The sampling location for the TSI-SMPS and NPS measurements is 1 m away 297 from the cooking spot, which caused sudden changes in concentration. The cooking activity was continued for 298 approximately 8 min. The size distribution obtained by the NPS is shown every 1 s while the TSI-SMPS 299 measurement provides one size distribution every 2 min (total 6 successive measurements). Therefore, the TSI-300 SMPS analysis provides discontinuous size distribution by time. Figure 9(c) shows the particle concentration at a 301 peak particle size for each measurement of the TSI-SMPS and NPS. From the NPS measurements during the 302 cooking activity, particle concentrations varied significantly. Relatively low particle concentrations were observed 303 approximately 180 s after the beginning of the activity, and then several peaks were observed until the end of the 304 event. Like these experiments, size distribution data obtained every 1 s by the NPS can be informative in various 305 applications.

306

Figure 9

307 **5 Conclusion**

308 In this research, we developed and evaluated the performance of a new Nano-particle sizer (NPS) that measures 309 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 310 311 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 312 313 normalized size distribution. The results were used to calibrate the NPS raw data to derive the real particle number 314 concentration and size distribution. The NPS was compared to the TSI-scanning mobility particle sizer (TSI-315 SMPS) for steady-state and unsteady particle concentrations using NaCl and Ag particles. The size distributions 316 obtained by the NPS under the steady-state condition agreed with the results from the TSI-SMPS. For the unsteady 317 particle size distribution with fast-changing particle concentration, the NPS was found to be superior to the TSI-318 SMPS in terms of measurement speed. However, there remains a needed improvement. During the NPS 319 measurements, we experienced electrical breakdown when the applied voltage was approximately 4000-5000 V. 320 Therefore, to improve the NPS system for wider size range classification, further optimization is required. From 321 the findings in this study, we believe that the NPS can be a promising instrument providing comprehensive 322 information on fast-changing concentration environments.

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325

326 **Competing interests**

327 Authors declare no conflict of interest.

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374 Captions

- 375 Figure 1. Schematic diagram of the NPS consisting of the MP-DMA including M-CPCs: (a) the geometry of the
- 376 MP-DMA and flow paths; (b) the details of the 12th home-made CPC; (c) the M-CPC module.
- 377 Figure 2. Schematic diagrams of (a) the M-CPC, (b) MP-DMA and (c) NPS performance tests.
- 378 Figure 3. M-CPC performance: (a) activation efficiencies of 12 home-made CPCs; (b) concentration linearity
- 379 between the electrometer and M-CPCs.
- 380 Figure 4. Normalized concentrations of the classified particles though 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.
- 384 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:
- 386 evaporation generator (low temperature), (b) Ag particle: evaporation generator (high temperature), (c) NaCl
- particle: Collison 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.
- 391 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
- 394 second TSI-SMPS scanning data; (3) the NPS data for 240 s. The data were obtained at the NPS applied voltage
- 395 of 1000 V.
- 396 Figure 9. Size distributions measured by the (a) TSI-SMPS and (b) NPS during a cooking activity and (c) variation
- 397 of particle concentration at mode diameters. The NPS data were obtained at the applied voltage of 1000 V.

Sectional View of NPS

Section View "A-A"





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



400 Figure 1







403 Figure 2









409 Figure 4





412 Figure 5





414 Figure 6













MP-DMA voltage Port number 9 1 2 3 4 5 6 7 8 10 11 12 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 Mode diameter [nm] 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

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