Rapid measurement of sub-micrometer aerosol size distribution using a fast integrated mobility spectrometer

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A B S T R A C T

Rapid measurement of submicron particle size distributions enables the characterization of aerosols with fast changing properties, and is often necessary for measurements onboard mobile platforms (e.g., research aircraft). Aerosol mobility size distribution is commonly measured by a scanning mobility particle sizer (SMPS), which relies on voltage scanning or stepping to classify particles of different sizes, and may take about 1 minute or longer to obtain a complete size spectrum of aerosol particles. The recently developed fast integrated mobility spectrometer (FIMS) with enhanced dynamic size range separates and detects particles from 10 to ~600 nm simultaneously, allowing submicron aerosol mobility size distributions to be captured at a time resolution of 1 s.

In this study, we present a detailed data inversion routine for deriving aerosol size distribution from FIMS measurements of aerosols with a wide size range. The inversion routine takes into consideration the FIMS transfer function, particle penetration efficiency in the FIMS, and multiple charging of aerosols. The accuracy of the FIMS measurement is demonstrated by comparing parallel FIMS and SMPS measurements of stable aerosols with a wide range of size spectrum shapes, including ambient aerosols and aerosols classified by a differential mobility analyzer (DMA). The FIMS and SMPS-derived size distributions show excellent agreements for all aerosols tested. In addition, total number concentrations of ambient aerosols were integrated from 1 Hz FIMS size distributions, and compared with those directly measured by a condensation particle counter (CPC) operated in parallel. The integrated and measured total particle concentrations agree well within 6.5%.

1. Introduction

Real-time measurement of submicron aerosol size distribution is of great importance in understanding the health effects, nanomaterial functionality, and climate influences of aerosols (Dusek et al., 2006; Hu et al., 2017; Jiang, Oberdörster, & Biswas, 2009; Li, Ren, Biswas, & Stephen, 2016; Stott et al., 2000; Wang et al., 2002). Among the techniques for real-time measurement, scanning mobility particle sizer (SMPS) based on a differential mobility analyzer (DMA) (Knutson & Whitby, 1975) is most widely utilized. The DMA acts as a bandpass filter, allowing only particles within a narrow range of electrical mobilities, i.e., mobility sizes, to pass through and be subsequently measured by a detector, such as a condensation particle counter (CPC) or an electrometer. As the mobility size of the classified particles depends on the classifying voltage of the DMA, the number concentration of particles with different mobility sizes is measured by scanning or stepping the classifying voltage across the DMA electrodes. The size distribution is then derived using a standardized data inversion method, which takes into account the particle charging efficiency, DMA transfer...
function, and particle counter detection efficiency (Collins, Flagan, & Seinfeld, 2002; Stolzenburg & McMurry, 2008). The SMPS has been broadly used in atmospheric aerosol measurements (e.g., Wiedensohler et al., 2012, Wang, Flagan, & Seinfeld, 2003) and engineered particle characterizations (e.g., Haddad et al., 2016; Wang et al., 2015; Nie, Wang, & Biswas, 2017).

Due to the necessity to scan the classifying voltage, aerosol size distribution measurements by the SMPS often take about 1 min or longer, during which aerosol properties may change significantly, especially for the measurements of aerosols with rapid dynamics or onboard mobile platforms (e.g., research aircraft). Whereas faster size scans may be possible with fast response detectors (e.g., Shah & Cocker, 2005; Wang, McNeill, Collins, & Flagan, 2002), they lead to reduced counting statistics therefore increased uncertainty in the measurements, as a result of reduced sampling time for each size bin. Faster electrical mobility-based size distribution measurements are feasible with the electrical aerosol spectrometer (EAS, (Mirme et al., 1984)) or with instruments derived from the EAS such as the fast mobility particle sizer (FMPS, TSI 3091). However, the low sensitivity of the electrometer limits the applications of these instruments to aerosols of high concentrations (e.g., engine exhaust). In addition, the uncertainty associated with the unipolar charger and the limited number of size channels reduce the size resolution and the accuracy of the size distribution measurements with these instruments (Jeong & Evans, 2009). Fast measurements of aerosol size distributions are also possible by using optical particle counters (OPC), which measure particle sizes based on particle light scattering. However, OPC measurements are generally limited to particles with diameters larger than 100 nm and are strongly dependent on the refractive index of the particles (Li & Biswas, 2017), which is a function of the particle size, chemical composition, and morphology (Hering & McMurry, 1991), and is often not well known for ambient aerosol particles.

A fast integrated mobility spectrometers (FIMS) was developed to measure aerosol size distributions with 1 s time resolution while maintaining good counting statistics and size resolution (Kulkarni & Wang, 2006a; Olfert & Wang, 2009). The original FIMS employs a constant electric field to spatially separate particles of different mobilities, subsequently grows the separated particles via the condensation of alcohol vapor, and finally captures the grown particle locations and concentration via imaging by a high-speed CCD camera (Kulkarni & Wang, 2006a, 2006b; Olfert, Kulkarni, & Wang, 2008). The particle locations are directly related to particle mobility sizes, and the number counts of particles within mobility bins are correlated with particle size distributions (Olfert et al., 2008). The dynamic size range of the original FIMS was significantly increased by replacing the constant electric field with a spatially varying one, such that particles with diameters ranging from 8 to 600 nm can be simultaneously measured. The FIMS employing the spatially varying electric field was characterized and shown to have a good sizing accuracy, near 100% detection efficiency for particles with diameters greater than 8 nm, and a mobility size resolution generally comparable to that of a traditional SMPS system (Wang, Pikridas, Spielman, & Pinterich, 2017; Wang et al., 2017). Further development of the FIMS also included the activation of the separated particles using the condensation of water vapor instead of alcohol vapor (Pinterich, Spielman, Hering, & Wang, 2017; Spielman, Hering, Kuang, & Wang, 2017) and the application of FIMS in aerosol hygroscopicity measurements (Pinterich, Spielman, Wang, Hering, & Wang, 2017).

In this study, we systematically examine the accuracy of aerosol size distributions derived from the measurements of the heptanol-based FIMS employing a spatially varying electric field (Wang, Pikridas, et al., 2017; Wang et al., 2017). A data inversion routine, which takes into account the aerosol charging efficiency, aerosol penetration efficiency, and the FIMS transfer function is presented. The FIMS and a traditional SMPS were operated in parallel to measure various laboratory generated and ambient atmospheric aerosols. The aerosol size distributions, together with the integrated total concentrations obtained from the two instruments are compared. In addition, the total particle concentration integrated from 1 Hz size distribution measured by the FIMS was compared with that directly measured by a CPC.

2. Material and methods

2.1. FIMS data inversion

2.1.1. FIMS setup

The detailed geometry and working principle of the FIMS with a spatially varying electric field were presented earlier by Wang et al. (2017) and Wang, Pikridas, et al. (2017). Here we adopt the same coordinate system as in Wang, Pikridas, et al. (2017). The FIMS consists of three sections in sequence: a mobility separator, a condenser, and a detection region, which share identical cross-section dimension \((x \times y = 1.12 \times 12.7 \text{ cm})\). After being introduced into the FIMS mobility separator via an entrance slit, charge-conditioned particles follow different trajectories based on their electrical mobilities in the separator and the vertical velocities brought by the heptanol-saturated sheath flow in the vertical \((z)\) direction. As a result, at the exit of the separator, charged particles are spatially separated according to their mobility size. These spatially separated particles then grow into micrometer droplets in a supersaturated environment, created by thermal electric cooling of the condenser. In the detection region, the grown droplets are illuminated by a sheet of laser that covers the entire channel cross section (i.e., \(x-y\) plane), and imaged by a high-speed CCD camera. Because the condenser is grounded; the particle position within the channel cross-section (i.e., \(x\) and \(y\) positions), which is related to particle mobility size, remains unchanged after the particles exit the mobility separator. Compared to the previous designs of the FIMS (Kulkarni & Wang, 2006a, 2006b; Olfert & Wang, 2009; Olfert et al., 2008), the current FIMS employs a new electrode that creates an electric field with strength varying drastically along the \(y\) coordinate, such that particles of a wide size range (i.e., 8–600 nm) can be simultaneously classified and detected. The CCD camera (Pantera TF 1M30, DALSA Inc., Waterloo, Canada) records 8-bit grayscale images of the illuminated particles and the channel walls at a frame rate of 10 Hz. The procedure to derived size distribution from FIMS measurements is described in the following sections.
2.1.2. Converting particle locations to instrument response diameters

Fig. 1a shows an example of particle positions in the x-y plane (i.e., cross section of the rectangular channel) retrieved from the recorded images. Only particles detected within the viewing window (i.e., $0.2a \leq x \leq 0.8a$, $-3.5\text{ cm} \leq y \leq 3.5\text{ cm}$, where $a = 1.12\text{ cm}$, and represents the gap between ground and HV electrodes) at the center of the channel cross section are used to derive aerosol size distribution, in order to avoid the edge effects of the electric and flow fields, and to maintain a mobility resolution above 4 (Kulkarni & Wang, 2006a, 2006b). To facilitate the derivation of size distribution from the FIMS measurements, we introduce instrument response mobility ($Z_p^*$) following the same approach as in Kulkarni & Wang (2006a) and Wang, Pikridas, et al. (2017). Basically, for a non-diffusing particle entering the separator along the central inlet flow streamline, the instrument response mobility at the x-y position where the particle is detected is the same as the particle mobility $Z_p$. Fig. 1b shows the map of the instrument response mobility ($Z_p^*$) in the channel cross section. The value of $Z_p^*$ was derived from simulated trajectories of singly charged, non-diffusing particles introduced at 1000 initial y positions evenly spaced from $-4.0$ and $4.0$ cm along the central aerosol flow stream line (Wang, Pikridas, et al., 2017). At each initial y position, particles of 2500 different diameters logarithmically distributed from 4 to $900\text{ nm}$ were introduced, and their positions at the exit of the separator were determined by simulating particle trajectories using the Langevin equation (Friedlander, 1977), assuming no Brownian diffusion. We note that for non-diffusing particles, their x-y positions at the separator exit are identical to those detected following condensational growth inside the condenser, where no electric field is applied. Due to the influence of the Brownian diffusion and the fact that particles enter the FIMS separator along different aerosol flow streamlines, particles with mobility of $Z_p$ may not be detected with the same instrument response mobility (similar to the working principle of the DMA). Therefore, the transfer function of the FIMS needs to be taken into account during the data inversion process as described in Section 2.1.3. Fig. 1c shows a color plot of the instrument response diameter ($D_p^*$) derived from $Z_p^*$ by using the Stokes-Millikan equation (Friedlander, 1977)

$$Z_p^* = eC/3\pi\mu D_p^*,$$

(1)

assuming singly charged particles, where $e$ is the elementary unit of charge, $C$ is the Cunningham slip correction factor, and $\mu$ is the dynamic viscosity of the carrier gas, which is a function of temperature and pressure during the measurement. In one of the first steps of the data inversion, the detected particle positions retrieved from each image are converted to $Z_p^*$ using the simulated $Z_p^*$ map (i.e., Fig. 1b), then to $D_p^*$ by using the above Stokes-Millikan equation.

Due to the parabolic flow profile, the particle residence time inside the FIMS (i.e., the time between particles entering the FIMS separator and being detected) varies with particle mobility. The residence time of each detected particle was derived using the same approach as described in Olfert et al. (2008). The time when particles arrive at the FIMS separator was then calculated by subtracting the residence time from the time of detection, and grouped into time intervals. In this study, we used time intervals of 1 s to derive aerosol size distribution at a time resolution of 1 Hz. Particles detected during each 1-s time interval was further grouped into

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**Fig. 1.** (a) The FIMS camera-recorded locations of 10–300 nm monodisperse particles within the viewing window after mobility separation and growth. The monodisperse particles were generated by classifying atomized (NH$_4$)$_2$SO$_4$ particles with a DMA at sheath-to-aerosol ratio of 10:1 (Wang et al., 2017). (b) and (c) show maps of instrument response mobility ($Z_p^*$) and diameter ($D_p^*$) derived from particle trajectories and positions at separator exit simulated using the Langevin equation.
instrument response diameter channels with the channel boundaries \((D_{p,i-1/2}^* \text{ and } D_{p,i+1/2}^*)\) evenly spaced on a logarithmic scale. The response of FIMS for the \(i^{th}\) \((i = 1, 2, 3, \ldots, I)\) channel \((R_i)\) represents the number of particles detected between boundaries \(D_{p,i-1/2}^* \text{ and } D_{p,i+1/2}^*\) during the time interval. In this study, 50 size bins \((i = 50)\) were used to cover a size range of 10–600 nm. The responses \((R_i \text{ to } R_{50})\) were then used to calculate the aerosol size distributions.

### 2.1.3. Inversion of aerosol size distribution from FIMS responses

For a given time interval, the response of the FIMS \((R_i)\) can also be derived from particle number size distribution. Let \(n(D_p) = \frac{dN}{d \log(D_p)}\) denote the particle number size distribution, the number concentration of particles with diameters between \(D_p\) and \(D_p + dD_p\) can be written as

\[
dN = \frac{dN}{d \log(D_p)} d \log(D_p) = n(D_p) d \log(D_p).
\]

The number of particles with diameters between \(D_p\) and \(D_p + dD_p\) that are detected within the \(i^{th}\) channel during the time interval of \(\Delta t\) is therefore given by:

\[
dR_i = Q \Delta t \int_{D_{p,i-1/2}}^{D_{p,i+1/2}} dZ_p^* \sum_{k=1}^{k_{\text{max}}} \eta_{\text{pen}}(D_p) \eta_{\text{lag}}(D_p, k) P(Z_p(D_p, k), Z_p^*) n(D_p) d \log(D_p)
\]

where \(Z_{p,i-1/2}^* \text{ and } Z_{p,i+1/2}^*\) are the instrument response mobilities corresponding to the \(i^{th}\) channel boundaries \(D_{p,i-1/2}^* \text{ and } D_{p,i+1/2}^*\), and were derived using the Stokes-Millikan equation. \(\eta_{\text{pen}}(D_p)\) is the fraction of particles that penetrate through the FIMS inlet line, which consists of a series of tubing, a laminar flow element, and narrow rectangular flow channels, including the aerosol inlet slit in the grounded electrode. The value of \(\eta_{\text{pen}}(D_p)\) was determined by experimental characterization together with numerical calculation based on the FIMS inlet geometry (Wang et al., 2017). \(\eta_{\text{lag}}(D_p, k)\) is the charging probability of particles carrying \(k\) charges, and was calculated using formula reported by Wiedensohler (1988). \(k_{\text{max}}\) is the maximum number of charges being considered, and in this study, \(k_{\text{max}} = 2\). \(P(Z_p, Z_p^*)\) is the transfer function of the FIMS, which describes the probability density of particles with mobility of \(Z_p\) measured by \(Z_p^*\). \(P(Z_p, Z_p^*)\) was derived from particle trajectories simulated using the Langevin equation, following the same approach described in Wang, Pikridas, et al. (2017). Based on Eq. (3), \(R_i\) can be derived by integrating the contribution from particles of different diameters:

\[
R_i = Q \Delta t \int_{D_{p,\min}}^{D_{p,\max}} d \log(D_p) \int_{D_{p,i-1/2}}^{D_{p,i+1/2}} dZ_p^* \sum_{k=1}^{k_{\text{max}}} \eta_{\text{pen}}(D_p) \eta_{\text{lag}}(D_p, k) P(Z_p(D_p, k), Z_p^*) n(D_p),
\]

where \(D_{p,\min}\) and \(D_{p,\max}\) are the lower and upper diameter limits of the aerosol size distribution. The integral in Eq. (4) can be rewritten as a sum over \(m\) diameter bins from \(D_{p,\min}\) to \(D_{p,\max}\), with the assumption that \(n(D_p)\) is constant within each size bin:

\[
N_i = \sum_{j=1}^{m} n(D_{p,j}) Q \Delta t \int_{D_{p,j-1/2}}^{D_{p,j+1/2}} d \log(D_p) \int_{D_{p,j-1/2}}^{D_{p,j+1/2}} dZ_p^* \sum_{k=1}^{k_{\text{max}}} \eta_{\text{pen}}(D_p) \eta_{\text{lag}}(D_p, k) P(Z_p(D_p, k), Z_p^*)
\]

where \(D_{p,j-1/2} \text{ and } D_{p,j+1/2}\) \((j = 1, 2, 3, \ldots, m)\) is the lower and upper bounds of the \(j^{th}\) size bin, \(D_{p,j} = (D_{p,j-1/2} - D_{p,j+1/2})/2\) is the geometric mean bin diameter, and \(n(D_{p,j})\) represents the concentration in the \(j^{th}\) size bin. Eq. (5) can be expressed in a matrix form as \(\mathbf{R} = \mathbf{M} \times \mathbf{n}\), where \(\mathbf{R}\) is an \(I \times 1\) array composed of \(R_i\) \((i = 1, 2, 3, \ldots, I)\) and \(\mathbf{n}\) is an \(m \times 1\) array composed of \(n(D_{p,j})\) \((j = 1, 2, 3, \ldots, m)\). The element of the matrix, \(\mathbf{M}\), is calculated by

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**Fig. 2.** A schematic diagram of the experimental setup for the comparison of parallel FIMS and SMPS measurements (top diagram) and parallel FIMS and CPC measurements (bottom diagram).
\[
M_{ij} = Q \Delta t \int_{D_p,j-1/2}^{D_p,j+1/2} \log(D_p) \int_{D_p,j-1/2}^{D_p,j+1/2} dZ^* \sum_{k=1}^{2} \eta_{pen}(D_p) \eta_{log}(D_p, k) P(Z_p(D_p, k), Z^*_p)
\]

(6)

For simplicity, the same boundaries were used for \(D^*_p\) channels and \(D_p\) size bins in this study, (i.e., \(l = m = 50, D_{p,i\pm1/2} = D^*_{p,i\pm1/2}, D_{p,\text{min}} = 10 \text{ nm}, \) and \(D_{p,\text{min}} = 600 \text{ nm}\). Deriving the size distribution function \(\bar{n}\) hence involves a data inversion process that calculates \(\bar{n}\) from \(\bar{M}\) and \(\bar{R}\).

The inversion of aerosol measurement data is a common problem in size distribution analysis (Kandlikar & Ramachandran, 1999). The most widely used technique was described by Twomey (1975), which is a non-linear iterative method. Twomey’s method was further improved by conducting data smoothing and adding criteria for completing iteration (Markowski, 1987). In this work, this revised Twomey method was used for calculating the aerosol size distributions. Data smoothing was not applied since it sometimes decreases the resolution of the derived aerosol size distributions, as reported by Markowski (1987) and Olfert, Kulkarni, and Wang (2008).

2.2. Experimental setup

Fig. 2 shows the experimental setup for the comparison of parallel FIMS and SMPS measurements of aerosol size distributions, and for the comparison of total number concentration integrated from the FIMS size distribution to that directly measured by a CPC. The tested aerosol samples included both ambient aerosols and laboratory-generated size-classified aerosols. Aerosol samples were dried by a diffusion dryer before being introduced to the SMPS and FIMS. The ambient aerosols were sampled near a roadside in the Brookhaven National Laboratory. The laboratory generated aerosols were produced by a constant atomizer (Model 3076, TSI Inc., Shoreview, MN, USA) nebulizing 0.1 g/l ammonium sulfate (> 99.0%, Sigma-Aldrich Inc., St. Louis, MO, USA)–water solution. The atomized aerosols were then size-classified by an electrostatic classifier (Model 3080, TSI Inc.) with a Long DMA (Model 3081, TSI

Fig. 3. Comparisons between the size distributions measured by the FIMS and SMPS for DMA-classified (a) 20 nm, (b) 40 nm, (c) 100 nm, and (d) 250 nm aerosols, and (e, f) two ambient aerosol samples. The size distributions were derived without applying the multiple-charging correction.
In this study, the size distributions derived from the FIMS measurements were compared with those measured by the SMPS. The SMPS (Model 3938, TSI Inc.), consisting of a long DMA (Model 3081, TSI Inc.) a butanol-CPC (Model 3772, TSI Inc.), was operated with a sheath flow rate of 3.0 l/min, providing a sheath-to-aerosol flow ratio of 10:1. Because the CPC flow rate is higher (1.03 l/min) than the monodispersed flow exiting DMA (0.31/min), the monodispersed flow was mixed with a stream of filtered air, maintained at 0.73 l/min by a mass flow controller (MKS Instruments, Rochester, NY, USA), before being introduced to the CPC. The SMPS was operated with a scan time of 300 s, a retrace time of 3 s, and a purge time of 10 s. A channel resolution of 32 channels/decade was used such that the total bin number of the SMPS (54 size bins in the range of 14.9–673.2 nm) was similar to that of the FIMS (50 size bins in the range of 10–600 nm). During the measurements of the DMA-classified aerosols, the size range scanned by the SMPS was narrowed accordingly to the size range of interest. The size distributions measured by the SMPS were directly exported from the Aerosol Instrument Manager® software.

The FIMS was operated at a sheath flow of 13.0 l/min and an aerosol flow of 0.25 l/min. The temperature difference between the condenser (cold) and the heptanol reservoir (room temperature) was maintained at 20 °C. There was no active control of the reservoir temperature, so during the operation, the reservoir and condenser temperatures varied slightly with the room temperature. The maximum and minimum voltages applied to the two terminal traces of the electrode were 6000 and 20 V, respectively (Wang, Pikridas et al., 2017). Size distributions at a time resolution of 1 Hz were inverted from the FIMS measurements, and averaged to SMPS scan time periods. The average FIMS size distribution was then compared with that measured by the SMPS.

Ambient aerosols were also measured by the FIMS and a CPC in parallel. During these measurements, a soft X-ray neutralizer was used in front of the FIMS inlet. The aerosol flow rate of the CPC was maintained at 1.03 l/min without further dilution. The total particle concentration was derived by integrating the FIMS size distribution between 10 and 600 nm. Due to the change of the aerosol neutralizer, the charge distribution of the aerosol sample also varied. Hence, the charging probability values for soft X-ray neutralizers (Tigges, Wiedensohler, Weinhold, Gandhi, & Schmid, 2015) were used for inverting the size distributions measured by the FIMS.

3. Results and discussion

3.1. Comparison of size distribution measurements by FIMS and SMPS

Figs. 3 and 4 show the size distributions inverted from the FIMS and SMPS measurements of the DMA-classified 20, 40, 100, and 250 nm aerosols, as well as ambient aerosol samples. The size distributions shown in Fig. 3 were calculated assuming particles can carry one charge only, while those shown in Fig. 4 were corrected for multiple charging. The results show excellent agreements between the size distributions measured by the FIMS and SMPS. The average particle sizes and total concentrations of the size distributions displayed in Fig. 4 were derived and further compared in Table 1. The relative differences between the SMPS and FIMS in average particle size and total concentration are within 6.5%.

In Fig. 3, the 100 and 250 nm particle size distributions also exhibit a secondary mode with a smaller mode size (Fig. 3c and d), resulting from the doubly charged 100 and 250 nm particles. This secondary mode becomes diminished, albeit not completely removed, after multiple charging is accounted for in the SMPS and FIMS inversion algorithms (Fig. 4d). This is possibly due to the different charger ion properties which resulted in somewhat different charging probabilities compared to those described in Wiedensohler (1998). This result also suggests that the multiple charge correction should always be applied when measuring particles larger than 100 nm.

3.2. Comparison of total number concentration measurements by FIMS and CPC

Ambient aerosol samples were also measured by the FIMS and a CPC in parallel (Fig. 1, bottom diagram). Fig. 5a displays the evolution of ambient aerosol size distributions measured by the FIMS near a roadside in the Brookhaven National Laboratory. The total concentration integrated from the 1 Hz FIMS aerosol size distribution is compared with the direct measurement by the CPC. The total number concentrations agree very well, with the difference within 5% (Fig. 5b). The high time resolution size distribution from the FIMS provides additional insights into the aerosol properties and processes. The size distribution was bimodal during the sampling period, showing an Aitken mode and an accumulation mode at approximately 50 and 130 nm (Fig. 5a). The size distribution also shows that the decrease of total aerosol concentration during 400–450 s was due to a lower Aitken mode particle concentration. Since the ambient aerosols were sampled by the roadside, the dynamics of the Aitken mode may be closely related with the generation and transport of engine-emitted aerosols.

The FIMS was deployed onboard the US Department of Energy Gulfstream-1 (G-1) research aircraft during the Observations and
Modelling of the Green Ocean Amazon (GoAmazon2014/5) experiment in the central Amazon basin (Martin et al., 2017). Fig. 6a shows an example of 1 Hz size distributions measured by the FIMS during the flight on March 17, 2014. The total particle number concentrations integrated from the FIMS size distribution agree well those measured by a CPC (Model 3010, TSI Inc.), with a difference generally below 5% (Fig. 6b). The fast measurement by the FIMS captured the spatial variation of the aerosol size distribution, and allowed clear identification of aerosols under background conditions, and those impacted by the pollution plume from Manaus (Fig. 6a). The rapid variation of size distribution, as observed when G-1 flew through the Manaus plume, cannot be resolved by traditional SMPS systems with a typical time resolution of minutes. The rapid measurements by the FIMS also helped identify that the vertical transport of Aitken mode particles from the free troposphere to the planetary boundary layer during precipitation events sustains the aerosol concentrations in the Amazon basin during the wet season (Wang et al., 2016).

![Graphs showing size distributions measured by SMPS and FIMS for different aerosols and ambient samples](image)

**Table 1**
The average particle sizes ($D_{p,mean}$) and total number concentrations ($N_{tot}$) derived from the size distributions measured by the SMPS and FIMS. The values were calculated from size distribution with multiple-charging correction applied, and correspond to the results in Fig. 4. Amb 1 and 2 represent the ambient samples 1 and 2, respectively. The relative differences (%) between the two measurements are also listed.

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<thead>
<tr>
<th>Test Aerosol</th>
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<th>FIMS</th>
<th>Difference (%)</th>
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<td>$D_{p,mean}$ (nm)</td>
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![Graph showing size distributions with multiple-charging correction applied](image)

Fig. 4. Same as Fig. 3 except that the size distributions were derived with multiple-charging correction applied.
4. Conclusion

In this study, the accuracy of aerosols size distributions measured by the FIMS is demonstrated by comparing parallel FIMS, SMPS, and CPC measurements. A data inversion procedure, which takes into account the FIMS transfer function, particle penetration efficiency, and particle charging efficiency is presented. Size distribution was derived iteratively from the FIMS measurements using an inversion matrix by the revised Twomey method. The size distribution shows excellent agreement with parallel measurement by a SMPS for stable aerosols with a wide range of size spectrum shapes, including ambient samples and size-classified aerosols. The total particle concentration integrated from the 1 s size distributions measured by the FIMS also agrees well with the CPC measurement, with a difference within 5%. The results show that the FIMS provides accurate measurement of aerosols size distribution with high time resolution, therefore greatly improves our ability in understanding rapid aerosol dynamics and characterizing aerosol properties using mobile platforms.

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