Characterization of drying bioaerosols with four size distribution measurement instruments in the context of airborne transmission

Xavier Lefebvre (xavier.lefebvre@polymtl.ca)
Polytechnique Montréal

Antonella Succar
Polytechnique Montréal

Emilie Bédard
Polytechnique Montréal

Michèle Prévost
Polytechnique Montréal

Etienne Robert
Polytechnique Montréal

Article

Keywords:

Posted Date: November 3rd, 2022

DOI: https://doi.org/10.21203/rs.3.rs-2207576/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License. Read Full License
Characterization of drying bioaerosols with four size distribution measurement instruments in the context of airborne transmission

Xavier Lefebvre¹,*, Antonella Succar¹, Emilie Bédard², Michèle Prévost², and Etienne Robert¹

¹Department of Mechanical Engineering, Polytechnique Montréal. 2500 Chem. de Polytechnique, Montréal, QC H3T 1J4
²Department of Civil Engineering, Polytechnique Montréal
*xavier.lefebvre@polymtl.ca

ABSTRACT

Measuring aerosol size distributions with precision is important to understand the transmission of infectious diseases causing pneumonia-like illnesses, including Legionnaire’s disease and COVID-19. Accurate size measurement can help guide the identification of risk mitigation strategies, a challenging task as the size of pathogen carrying particles can change due to evaporation. Using oil and water-based particles, we compare four measurement instruments that provide aerosol size distributions relying on different measurement principles; phase doppler anemometry, light scattering, electrical mobility and aerodynamic impaction. The size distributions obtained from the different measurement instruments are consistent for oil-based particles but differ significantly for particles experiencing evaporation. Experiments were conducted to quantify the differences in measurement caused by evaporation as well as the measurement losses. The size range covered by the four instruments is from 0.025 to 10 μm. Finally, an approach based on the complete drying of a salt solution is presented to obtain the size distribution at the source of the aerosol generation. Understanding of the infectious dose and the pathogen concentration at the source, the most dangerous size ranges of the aerosols can thus be determined, and actions can be taken to reduce the propagation of these particles.

Introduction

The ability of aerosols to carry pathogens has been known for over a century [1] and their role as a mean of infection has since been the subject of a sustained research effort. The ongoing COVID-19 pandemic has highlighted the need to improve our understanding of pathogen contamination to effectively prevent outbreaks [2]. Two distinct transmission routes are traditionally recognized in the literature: droplet transmission and airborne transmission. Droplet transmission occurs through comparatively large droplets with a typical threshold at 10 μm [3]. As these droplets quickly settle on surfaces because of gravity, they can only affect people near the source since deposited particles cannot be inhaled [4]. If inhaled, large particles are typically captured in the upper airways and are therefore less likely to be responsible for transmission of respiratory tract infections [5]. As for airborne transmission, smaller droplets settle slower or not at all depending on the air currents present in indoor environments [6]. Consequently, they remain in the air for a longer time and can reach a very small but stable diameter through evaporation. These droplets, dependent upon environmental conditions will lose their water content and leave behind a small nucleus made of the solids contained in the droplet when it was formed, including potential pathogens [7]. Such sub-micron droplet nuclei can travel further from the source, effectively remaining airborne until actively removed through ventilation systems [8], and penetrate deeper in the airways when inhaled. A significant difficulty when dealing with bioaerosols is that they are for the most part water-based. As soon as a droplet separates from the bulk liquid where it is formed, it starts to shrink through evaporation, unless relative humidity is 100%, in which case it grows [9]. Measuring aerosol size distribution accurately is therefore essential to identify if particles are small enough for deposition in the respiratory tract, to improve the understanding of particle dispersion in ambient air and to assess the performance of aerosol control procedures or tools. More generally, size distribution measurements can be used to characterize a wide range of complex multiphase flows. Aerosol size distribution can be measured with a wide variety of instruments [10], which detect particles in different size ranges and rely on different operating principles. The large array of available size distribution measurement instruments covers particles sizes from 0.022 μm to more than 100 μm [11]. Some instruments are portable for field measurements while others are too large or sensitive to deploy outside of the laboratory. Their price range also vary considerably depending on their working mechanism. The instrumentation currently used in experimental studies of bioaerosols consists mainly of
impactors and filters [12]. These choices are motivated by the need to obtain cultivable samples and the generally low biological content in air requiring large volumes to be sampled. However, filters and impactors provide only limited information on the size of the particles carrying pathogens in bioaerosol, and only do so when implemented in cascade arrangements that are cumbersome to use. This information is critical as it controls the settling and deposition dynamics of the particles. Recent investigations have demonstrated that even small differences in particle size can have a major effect on the behavior of pathogen-carrying particles in indoor air, in the airways and in aerosol treatment systems [13] [14] [15], especially in the sub-micron range.

This has motivated several research groups working with bioaerosols to use precise analytic tools available in the field of aerosol science, that can quickly and conveniently provide detailed particle size distributions [16] [17]. These instruments are however currently unable to capture culturable samples and must therefore often be used in conjunction with sampling tools such as filters and impactors. Risk assessment models rely on the estimation of particle distribution in infection studies. These infections studies are subjected to the same uncertainties regarding the size, the number and the stability in time of wet particles. As different physical phenomena are at the heart of the measurement principle employed by these instruments, a strong research need remains to provide guidelines for meaningful interpretation of such experimental data.

In this study, a transversal comparison between instruments using different measurement techniques is presented. Two optical instruments were used: a Phase Doppler Anemometer (PDA) that detects single particles going through a measurement volume without sampling air [18], and a portable optical aerosol spectrometer (OAS) (GRIMM mini LAS 11-R) [19]. Detailed sub-micron size distributions were also obtained through the electrical mobility of particles using a Scanning Mobility Particle Sizer (SMPS) [20]. Finally, a Cascade Impactor was used to obtain the size distribution through the aerodynamic diameter of particles [21], a method used in most most bioaerosol studies.

Several laboratory and field studies provide comparisons of dry aerosol characterization, but scarce information is available on water-based aerosols. The SMPS being the gold standard for sub-micron particle size distribution measurements, it is often used as a comparison basis for other instruments [22]. Sousan et al. [23] compared the SMPS with an OAS for particle matter and found that the OAS overestimated the concentration of particles smaller than 0.5 \( \mu m \), but the instruments provided similar concentrations for particles larger than 1 \( \mu m \). Mass-based concentrations were more similar but cannot be applied to water-based aerosol because of evaporation [24]. Moore et. al [25] compared different spectrometers against a reference SMPS and suggested that disparities between optical instruments could be due to the different optical properties of particles. Since the size of water-based droplets is changes dynamically, optical properties could also vary across the size range of interest, especially with the presence of solids in the droplets. A PDA system was found to account for particles changing size when it was compared to image analysis for spray particles, despite a tendency to underestimate the size of particles [26]. Dodge [27] compared multiple instruments for sprayed droplets including a PDA system. His general conclusion is that only instruments using the same measuring principles yield matching results and differences in average size distribution can be as big as a factor of 5. Finally, Vo et. al [28] compared different portable optical size spectrometers, suggesting that portable instruments are as precise as their larger counterparts, while providing the advantage of being sturdier and easier to transport.

In a sampling instrument such as a cascade impactor or an OAS, evaporation can affect particle size from its creation to its sampling point and throughout the sampling line until the detector. For non-sampling instruments such as the PDA, only the evaporation between the creation site and the measurement volume needs to be taken into account. The presence of dissolved salts, bacteria or viruses in liquid droplets suspended in air has a major influence on the mass exchange occurring at the droplet interface, between the air and water. The consequence of the presence of salt from a thermodynamic point of view is that unless the relative humidity is very low, some water will remain in the droplet nuclei. This remaining water contributes to more favorable conditions for virus transmissions compared to completely dried droplet nuclei because the survivability rate of viruses is increased [30]. In this work, we use this phenomenon to provide a reliable comparison between instruments relying on different operating principles. By completely drying salt water-based aerosols using a gas column desiccator and measuring the size distribution of the NaCl nuclei, the size of droplets as they were generated can be calculated.

Comparing results from size distribution measurements of wet aerosols from instruments using different operating principles is challenging due to the nature of the instruments. As such, actionable data to assess risks is needed to optimally characterize bacteria-laden aerosols. Four different experimental approaches were used for this investigation. Aerosol measurement instruments were compared in the first two experiments using a flow of oil-based and water-based particles, suspended in
air. In the third experiment, the size distribution of water-based aerosols was measured with only the OAS and the PDA to study the effects of evaporation in the sampling line of the measurement. The last experiment involved measuring the dried size distribution of particles generated from a saline solution to then calculate the initial diameter of the particles.

Results and Discussion

Instrument comparison

The results of the comparison of the GRIMM, the SMPS, the PDA and the Cascade Impactor are shown in figures 1 and 3 for DEHS and water-based aerosols, respectively. The DEHS distribution covers the 0.01 to 10 \( \mu m \) and therefore includes respirable airborne particles. For the water-based distribution, the studied size range is from 1 to 20 \( \mu m \) as droplets generated with the spray gun were significantly larger. Repeatability was assessed with a triplicate for every measurement, with the standard deviations illustrated with the shaded areas. The SMPS and PDA distributions are composed of 100 size bins, while the GRIMM and the Cascade impactor only produced size distributions with 9 and 6 bins, respectively, for the DEHS measurements. The size resolution improved slightly for water-based aerosols with respectively 11 and 7 bins for the latter two instruments.

![Figure 1. Comparison of the four instruments with DEHS aerosols. Results are averaged over three measurements. Shaded areas show the standard deviation within repeated runs.](image)

Figure 1 compares the measurement instruments for DEHS aerosols. As much as \( 7 \times 10^7 \) particles per liter of air were observed. Because of the fine spatial resolution of the PDA, small localised perturbations in the flow, caused by the aerosol generators caused large changes in the distribution, but measurements demonstrated excellent repeatability above 0.5 \( \mu m \). Since the wavelengths of the lasers used in the PDA are approximately 0.5 \( \mu m \), the measurements for smaller particles lack precision and are illustrated as dashed line. The cascade impactor measurements showed the highest variability in concentration. For the SMPS, greater variability was observed close to the peaks of the distribution. The GRIMM displayed the most repeatable measurements because since measurements are made every 6 seconds and therefore, more data can be averaged over the measurement period. On the contrary, the PDA and SMPS measurements are made every minute, which explains the larger observed variability since the possibilities for perturbations in the flow are more likely the longer it takes for the measurement.
The peaks in the size distribution coincide for all instruments at approximately 0.3 µm, although the SMPS peak is slightly shifted towards smaller particles. The cascade impactor had the lowest number of size bins in the studied range, making the distribution less accurate, which could explain the shift of the peak towards larger particles. Sizes smaller than 0.25 µm are not shown for the cascade impactor because the measured mass was too small, causing large errors in the number concentration conversion. As with the cascade impactor, the GRIMM measured only approximately ten points in the studied size range, explaining why the distribution does not span the full range. The SMPS measurements delivered the expected lognormal curve and generally detected more particles than other instruments, probably because the size of the aerosols was in the preferred size range of the instrument. Although the DEHS particles were on the lower size limit of the measurement ranges of the instruments, the distributions from all four instruments show good qualitative agreement.

The advantage of using the PDA for number density and size distribution measurements is that no losses can occur within the sampling line. With the small inlet tube of the GRIMM and SMPS spectrometers, some particles are lost by impaction on the sampling tube walls, a phenomenon that can be quantified by comparing the concentration measurements obtained from the different instruments. Our results reveal that this effect appears only significant with the GRIMM, as shown by the generally lower particle concentration measured using this instrument. As shown in Figure 2, for particles larger than 0.25 µm, differences in the measurement were at least 70%, and from 0.8 µm, reached close to 100%, revealing that larger particles are less likely to be detected by the GRIMM. In addition to the loss of particles in transit, this important difference in the measured concentration between the GRIMM and the other instruments can also be caused by the different operating principles. However, as the mode and shape of the distribution are qualitatively in agreement, the results provided by this highly portable instrument remain valuable, especially when measuring dry aerosols. Regarding the cascade impactor, each stage is associated with a D50 value and consequently, particles with a given size are collected with 50% efficiency. For the SMPS, near the D50 cut-off of the inlet impactor at 0.617 µm, the concentration measured is expected to be 50% of the real value. If doubled, the results of the SMPS near the upper limit of its size range would match those of the PDA, revealing excellent agreement between these instruments.

Figure 3 reveals that the agreement between instruments is not good for the size distribution measurement of water-based aerosols when evaporation is ongoing, although useful information can still be extracted on the size distributions. The larger particles generated with the spray gun allowed the drying particles to be measured at a more suitable size with respect to the detection ranges of the instruments. As much as 10 x 10^10 particles per litre were measured, which is significantly more than for DEHS aerosols.

The PDA system measured larger particles than the other instruments. This is expected as particles don’t have to travel through a sampling line to be measured with the PDA and evaporation is therefore minimal compared to the other instruments. The cascade impactor reported more particles than the other instruments, with the shape of the distribution close that of the SMPS, with the exception of a dip at 0.15 µm. The assumptions used in the interpretation of the cascade impactor data could cause the overestimation of the number concentration. Because water-based aerosols dry quickly after being generated, they are less likely to remain spherical and since they lose water, their density changes over time, increasing after evaporation. The SMPS measured almost the same concentration as the PDA, but with smaller diameters, revealing that differences were dominated by the evaporation of particles during the sampling and the measurement process.

Again, the GRIMM measured significantly fewer particles than the other instruments. The difference increased up to 5 orders of magnitude compared to the other instruments, questioning the use of the GRIMM for wet particles. It can be assumed that larger particles were lost in the sampling line, and that evaporation plays a significant role in the measurement. The GRIMM and the SMPS therefore provide very different size distributions for two distinct reasons; first, because evaporation does not affect both instruments equally, with the longer transit time and dilution with the sheath flow in the SMPS likely causing particles to shrink further. Second, because in the GRIMM, the larger particles appear to suffer significant losses while in transit in the sampling line. The likelihood of particle loss through impaction in the sampling line can be characterized by the dimensionless Stokes number, which is the ratio of the characteristic time of a droplet t₀ to a characteristic time of the flow. Knowing the particle density ρₚ, the diameter of the particles dₚ, the fluid dynamic viscosity µₕ, and the flow speed u, the Stokes number in the case of droplets is defined as: 
Figure 2. Sampling losses observed with the GRIMM compared to the other three instruments. Results are averaged over three measurements. Shaded areas show the standard deviation within repeated runs.

\[ Stk = \frac{t_0 u}{d_p} \]  

where 

\[ t_0 = \frac{\rho_p d_p^2}{18 \mu_g} \]  

For large particles, the Stokes number is therefore large and impaction on the sampling line walls is more likely to occur because of the inertia of the particles. The flowrate is 1.2 L/min in the sampling line of the GRIMM, 2.0 L/min for the SMPS and 10 L/min for the cascade impactor. For a 0.5 \( \mu \)m particle, this results in a Stokes number of \( 9.8 \times 10^{-3}, 4.1 \times 10^{-3} \) and \( 2.02 \times 10^{-3} \) respectively inside the sampling lines of the GRIMM, the SMPS and the cascade impactor. Essentially, the Stokes number for GRIMM measurements is more than twice that of the SMPS and is five times larger than that of the cascade impactor, which explains the losses in the sampling line for larger particles with the GRIMM.

The observed differences, exceeding in some cases one order of magnitude, is significant but to be expected. Agreement between measurements for the DEHS aerosols was much better. Disparities for water-based aerosols revealed the critical role played by evaporation. Even though it is more apparent for water-based aerosols from the more significant differences between instruments, losses in sampling lines occur with both DEHS particles and water-based particles. The differences here are exacerbated by the fact that these losses are strongly size dependant, with the smaller water-based particles much more likely to be lost from impaction on the tube walls as the Stokes number is higher for larger particles.

**Aerosol Drying**

The results above clearly demonstrate that sampling losses and evaporation must be considered to estimate the real size of water-based bioaerosols as they were created, and to properly assess the threat they pose. This was investigated here using both
Figure 3. Comparison of the four instruments with water-based aerosols. Results are averaged over a triplicate of the measurements. Shaded areas show the standard deviation within repeated runs.

Evaporation can occur in the sampling line as well as in the instrument itself. Interestingly here, the GRIMM measured a similar particle concentration to that of the PDA at its peak, revealing that evaporation was more significant than losses inside the tube which can be estimated at 15% in this configuration. Differences in the distribution range from 68% to almost 100% for the larger particles of the distribution which is not surprising since there are more losses in measurement for larger particles with the GRIMM. The difference in total particles measured could also be in part due to evaporation inside the tube, making the particle diameter smaller and thus undetectable by the GRIMM.
Figure 4. Comparison of the GRIMM and the PDA to show the effect of the sampling tube on the measurements. Results are averaged over triplicate measurements. Shaded areas show the standard deviation within repeated runs.

Figure 5 shows the size distribution of dried NaCl droplet nuclei obtained from the SMPS. The distribution measured with the SMPS without drying the aerosol in the dessiccation column (wet distribution), is also shown, as well as the size distribution calculated from the procedure described in the methodology to obtain the particle size as they were generated (source distribution). The concentration of the latter accounts for evaporation and losses in measurement. However, as it was calculated rather than measured, it might not be representative of the real concentration size distribution at the source. As expected, the measured particles are much smaller than the particles at the source since evaporation occurs. The maximum number of particles measured was approximately $2 \times 10^9$ particles per liter of air for the dried distribution and the maximum was approximately three times more for the wet distribution, revealing significant losses in the drying process. Part of those losses most likely occur inside the desiccation column, as these were previously shown to be significant for small particles \cite{31}. The undried distribution measured with the SMPS had a maximum concentration at a diameter 2 times smaller than the dried distribution. These observations reveal that the desiccator-linked losses are more significant for smaller particles, perhaps through diffusion and electrical effects in the drying media.

The dried and wet distributions cover the same size range, are both essentially under 1 $\mu$m, and are cut off at the upper detection limits of the SMPS. The spray gun used to generate the aerosol yields a very broad size distribution, with small particles generated giving the distribution an elongated shape towards smaller sizes. Particles at the source ranged from 0.65 $\mu$m up to 10 $\mu$m and covered a broader range of particles than the dried and wet measurements. The dried distribution is more skewed towards larger particles compared to the wet distribution, because evaporation is slower for larger particles. Consequently, particles remain larger for longer periods of time in the sampling line, which would explain why the majority of the particles are measured around 0.1 $\mu$m in the wet distribution and around 0.2 $\mu$m in the dried distribution. Calculations of the source size distribution did not account for particles larger than 10 $\mu$m, but it can be assumed that particles much larger are generated by the spray gun. Accounting for this, 90 % of the generated particles are larger than 1 $\mu$m and once measured, all the particles are submicron. In the context of airborne transmission, this implies that the generated particles are large enough to trap large quantities of pathogens and lose water through evaporation until small enough for deep deposition in the respiratory tract. This configuration therefore allows for proper risk assessment of the aerosols generated from a given source.
Figure 5. Size distribution from the SMPS with dried NaCl aerosols and the calculated size distribution at the source. Results are averaged over triplicate measurements. Shaded areas show the standard deviation within repeated runs.

Significance of Results
The results presented demonstrate the challenges, but also the opportunities of combining aerosol concentrations and size distributions obtained from different instruments. Table 1 compares multiples characteristics of the size distribution measurement instruments as well as their advantages and limitations. The PDA is designed for spray measurements where the particle velocity and size distribution can be measured precisely. However, the determination of accurate particle concentration in an aerosol flow constitutes a challenge that requires precise measurement of the measurement volume illuminated by the lasers. The instrument also loses precision under 0.5 µm because of the limitation intrinsic to the wavelength of the laser. Alternatively, the SMPS is considered the gold standard for submicron particle measurement, yet it is susceptible to water-based particle drying and it cannot measure particles larger than 1 µm. Consequently, for applications such as airborne transmission, a critical size range potentially dangerous for transmission remains inaccessible. Moreover, both the PDA and the SMPS are complex to operate, expensive and difficult to move outside of laboratory settings. In contrast, the GRIMM and the cascade impactor are lighter, easy to handle and transportable. The cascade impactor measures the weight of the particles whereas the other instruments measure particle count. It also is the instrument with the lowest number of size bins, therefore providing less information about the particles than other instruments for the same size range. As for the GRIMM, a significant amount of particles are lost to the sampling tube walls as well as to evaporation, and it has limited precision for submicron particles. It must also be sent for calibration regularly. It has the major advantage however of providing a full size distribution every 6 seconds enabling complex parametric studies on aerosols to be conducted quickly. Furthermore, measurement time and sampling volume rate must be taken into consideration in the measurement instrument selection, especially when sampling in situ, as the measured flow may be suspect to atmospheric perturbations during sampling. Overall, precise size distribution measurement of a water-based aerosol flow, both in terms of concentration and size resolution, can only be obtained by combining instruments. The data presented here allows the sampling losses and the effects of evaporation to be estimated when only one instrument is available. Moreover, if samples are needed for further analysis of the particles, filters of impactors are used. Combining with data from spectrometers bring value, but must be done carefully.

Furthermore, as shown by our results, aerosol measurements are highly dependant on evaporation. Particles can only be
measured at a specific instant which doesn’t take evaporation into account since the size of particles is constantly changing. Having a way to obtain information on particles without having to consider evaporation is paramount to airborne transmission. Using the dried size distribution and microbial concentrations expected in various situations, one can calculate an estimate for the average number of pathogen units per droplet. These estimates can then be used as inputs to exposure models to identify the situations where aerosol-mediated transmission is plausible. The presence of aerosols in the size ranges that can be inhaled and contain microbial contaminant can be determined, and actions can be taken to reduce the risks associated.

Work is still needed on the evaporation of bioaerosols. The transfer rate of pathogens (bacteria, viruses, etc.) or of solids, from the bulk liquid to the aerosol needs to be determined. Then, the fate of the particles depending on their size must be studied for specific transmission scenarios. By quantifying the evaporation time and comparing it to the time needed for droplets to reach the ground, a critical droplet diameter can be defined depending on the droplet content, the environmental conditions and the pathogen of interest. This makes it possible to go beyond the traditional droplet/airborne dichotomy that clearly has limitations to describe the complex nature of aerosol-mediated transmission. Additional experiments must also be carried out to figure out how measurements from the studied instruments differ for particles that are not spherical, such as certain bacteria, and if measurements can be corrected if needed.

**Methods**

**Aerosol Generation**

Instruments were first compared through the measurement of particles generated by a TOPAS GMBH model ATM 221 aerosol generator that can be operated with water, a saltwater solution or Di-Ethyl-Hexyl-Sebacate (DEHS) oil. In all cases, the particles are in the sub-micron range and the distribution is reproducible. The size distribution generated from this type of aerosol generator fluctuates by a maximum of 2.5% over the course of several hours of use [32]. DEHS aerosols, with a density of 0.9 g/cm³, are spherical and dry very slowly so their size distribution can be considered constant. During operation, the aerosol generator is connected to a mass flow controller (Hastings HFC 202), enabling control over the output flow rate of aerosol. The second aerosol production approach is based on a spray gun (1.0 mm Mini HPLV Air Spray Gun, Neiko) operated with either tap water or 10% m/m NaCl dissolved in tap water with a density of 2.16 g/cm³. For saltwater-based measurements, aerosols were completely dried with a gas desiccator column filled with silica gel beads (Model DDU 570, TOPAS GMBH). The spray gun, operated with 6 bars of pressured air, generated larger particles compared to the TOPAS aerosol generator and was better suited to cover the whole size range of the measurement instruments that were used in the study.

**Instrumentation**

**PDA**

The PDA (PDA/LDA system, Dantec Dynamics, Skovlunde, Denmark) technique is an extension of laser Doppler anemometry and is based on phase Doppler principles. The measurement volume is defined by the intersection of two pairs of laser beams of 532 and 561 nm. As single particles flow through the measurement volume, light is scattered from the interference patterns created by the two laser beam pairs [18]. Size distribution can be accurately measured, as well as particle velocity. Since the measurement volume is very small, measurements are considered punctual, a characteristic separating the PDA from the other instruments, making it ideal for the measurement of spray particles [33]. The measurement size range is from 0.5 µm to 8000 µm for the instrument used in this study.

The settings used in the study, shown in table 2, were selected to ensure that the maximum number of particles were measured by the PDA, without saturating the detectors. Both lasers power was set to the maximum 300 mW and the gain was set to 20 dB for both detectors. The sensitivity of the detectors, which is adjusted with voltage, was the driving factor in the number of particles measured. The detector voltage was set to 1300 V when using DEHS aerosols and to 900 V when using water-based aerosols. When salt was added, the sensitivity was set to 1100V. Repetitive measurements were taken every 60 seconds for 5 minutes and averaged in a single size distribution profile.

**OAS**

The OAS (MiniLAS model 11-R, GRIMM Aerosol Technik Ainring GmbH & Co, Ainring, Germany), hereafter called the GRIMM, is an optical instrument that measures the light scattering of single particles with a diode laser to obtain the size distribution of aerosols [19]. The GRIMM is usually employed for atmospheric measurements. However, when applied to an aerosol flow size distribution measurement, it is common to connect a sampling tube to the inlet of the instrument [34] [35]. For this study, a 1.5 m sampling tube was connected at the inlet of the instrument for particles measurements. The inlet flow was 1.2 L/min. Particles with a size ranging from 0.25 to 32 µm were measured. The instrument requires knowledge of the
particle density for mass distribution measurement. Repetitive measurements were taken every 6 seconds for 5 minutes and averaged in a single size distribution.

**SMPS**
The Scanning Mobility Particle Sizer (model 3938 L76, TSI Incorporated, Shoreview, MN, USA) used in the study, hereafter called SMPS, consists of an electro-static classifier (EC, model 3082), a long differential mobility analyser (DMA model 3081A) and an ultrafine condensation particle counter (CPC model 3736), which is butanol-based [20]. In the DMA, the particles are drawn to an electrically charged rod at different rates as a function of their size, which only lets one particle size go through. The condensation particle counter finally counts the particles of each size and creates the size distribution.

The electrostatic classifier (EC) was fitted with a 71 \( \mu m \) impactor nozzle, a flow rate of 1.2 L/min was drawn with a sheath flow rate of 2.0 L/min. The scanning time was 60 seconds, the retrace time was 6 seconds and the purge time was 10 seconds, which yields a total time of 76 s for each measurement. These settings result in a diameter measurement range of 22 nm to 671 nm. The samples were drawn through plastic tubes of 4 mm in diameter connected to a 2 m flexible silicone tube (TSI Conductive Silicone Tubing) at the inlet of the instrument. Results are presented with diffusion correction applied in the software. Repetitive measurements were performed every 2 minutes for a total of 3 scans, averaged in a single size distribution. The impactor nozzle was cleaned between every set of 3 scans.

**Cascade Impactor**
The aerosol flow was also measured with a Cascade Impactor (Dekati® Low Pressure Impactor DLPI+). Particles with sizes ranging from 0.16 to 10 \( \mu m \) were collected on 14 different stages based on their aerodynamic diameter, with a sampling flow rate of 10 L/min for 30 minutes. The size associated to each stage is a D50 value, implying that each particle with a given size or larger are collected with 50% efficiency. The larger particles have more inertia and are collected on the first stages while the smaller particles have less inertia and are found on the lower stages. The samples were drawn through a flexible plastic tube with a diameter of 12.7 mm and a length of 0.6 m. Each stage was cleaned with ethanol and water before measurements, and weighed before and after measurements. Number size distribution was then derived from the mass of particles collected on each stage, assuming sphericity and constant density.

**Experimental Setup**
The objective of the experimental setup illustrated in Figure 6 was to dilute aerosols with clean air to produce a well mixed and low speed flow into a sampling chamber common to all instruments. The low flow speed into the sampling chamber results in equal velocities between the air around and inside the sampling tube, which minimizes divergence of the flow lines at the sampler inlet [36]. This isokinetic sampling prevents skewed size distribution measurements due to inertial effects of particles entering the sampling tube. The sampling chamber had a volume of 53 L and was made of plexiglass so that the optics of the PDA could measure particles through the side walls. Honeycomb material was fixed at the inlet of the mixing chamber as well as at its outlet to ensure a uniform flow. Finally, plastic tubes of 4 mm inner diameter fed the SMPS and the GRIMM. Another tube of 12.7 mm in diameter fed the cascade impactor. During our measurements, the temperature in the laboratory was in average at 25°C and the relative humidity was in average 42%.

Because of the different working mechanisms of the aerosol measuring instruments, the operating size range, as well as the detection and saturation limits are, different for each instrument. To enable the comparison of the size distribution measured by each instrument using the same aerosols, it was necessary to dilute with clean air to avoid saturation. For DEHS aerosols, the injection was done perpendicularly to the longitudinal direction of the tube and set by the flow controller at 4.2 L/min, for a dilution ratio of 1:5. The clean dilution air was injected from the other side of the tube, in the same manner as the aerosol. By injecting the aerosols and the air perpendicularly from the longitudinal direction of the tube, turbulence is created, and promotes the mixing of the two flows. To ensure that the flow is properly mixed and that a steady state concentration is reached, a tube of 1.83 m with an inside diameter of 0.0762 m was used between the aerosol inlet and the sampling chamber. Measurements were also started 3 minutes after flow initiation. Following every measurement, the chamber was drained by turning off the aerosol generator and injecting compressed air at 6.6 L/min for 3 minutes. For all experiments, the measurements were replicated three times to assess repeatability.

**Number Concentration**
To account for the fact that the number of size bins is not the same for every instrument, and that the bins are inequal in width, the y-axis on size distribution plots is normalized by dividing the number concentration of each bin by the logarithm of the bin width [23]. This results in a log-normal distribution. Some instruments such as the SMPS already provide a normalized
size distribution, but for the GRIMM, the Cascade Impactor and the PDA, the concentration had to be normalized through data post-treatment. While the GRIMM and the SMPS produce the size distribution by volume of air sampled, the PDA only counts the number of particles and the cascade impactor yields the cumulative mass of the particles collected on each stage. The concentration $N_C$ therefore needs to be calculated in the case of the PDA and the cascade impactor. For the latter, since the measurement was made over a period of 30 min and the flow is pumped at a constant rate of 10 L/min, the mass concentration was obtained for each stage by dividing the accumulated mass $M$ by the pumping rate $\dot{V}$ and the time of measurement $t$. The number concentration was then calculated assuming sphericity and constant density where $V_p$ is the volume of one particle of the measured size.

$$N_C = \frac{M}{\rho_p V_p \dot{V} t}$$  \hspace{1cm} (2)

For the PDA, the cross section of the measurement volume perpendicular to the flow represents the measurement surface needed to calculate the volume of gas covered by the laser. The ellipsoidal measurement volume provided by the supplier was verified by laser irradiance measurement [37], and confirmed $(2.6\,\text{mm} \times 153.1\,\mu\text{m})$. Assuming that the measurement volume is ellipsoidal in shape, the measurement surface could be derived. The speed of the flow through the measurement volume is known, the instrument also provides particle velocity, and the time of the measurement remains constant. The concentration $N_P$ is therefore obtained by dividing the counted particle number $N$ by the mean flow velocity $V_m$, the time of measurement $t$ and the measurement area $S$.

$$N_P = \frac{N}{SV_m t}$$  \hspace{1cm} (3)

**Aerosol Drying**

Aerosol generation from DEHS was best for instrument comparison since the particles only dry over several hours in ambient conditions. Additionally, NaCl water-based aerosols were used to study evaporation from particles. This problem was investigated by comparing the size distribution measured by the GRIMM and the PDA, using the spray gun to generate larger aerosol particles. To remove the complexity associated with uneven evaporation for size distribution measurement, the aerosols were completely dessiccated and measured from the sampling chamber with the SMPS. The SMPS was used because dried...
particles are smaller and in the right size range for measurement with this instrument. Knowing the concentration of salt in the water solution $C_s$ as well as the measured mass of salt $M_f$ and the quantity of particles for each size bin, the size distribution of the particles generated at the source $D_s$ was derived from simple calculations, assuming that the transfer rate of NaCl from the solution to the aerosols was constant. The size distribution was also measured without using the desiccator, for comparison, with $\dot{V}$ the sampling flow rate and $t$ the measurement time.

\[
D_s = \sqrt[3]{\frac{6V}{\pi}}
\]

where

\[
V = \frac{M_f\dot{V}t}{C_s}
\]

Conclusions

A transversal comparison is presented between four aerosol size distribution measurement instruments with different working mechanisms, realized in a flow of water and DEHS aerosols suspended in air. In general, instruments agreed when measuring particles that do not evaporate, but not when measuring water-based aerosols. For DEHS (oil-based) particles, all instruments detected comparable size distributions and concentrations with minimal differences, except for the GRIMM, which generally measured at least 70% less particles for sizes larger than 0.25 $\mu$m and up to 100% less particles throughout the distribution compared to other instruments. For water-based aerosols concentration differences affected all the instruments, reaching multiple orders of magnitudes.

Evaporation was the main factor explaining the differences observed between the measurements instruments. Throughout the size distribution, evaporation accounts for losses of 15 to almost 100% of the particles between measurements from the PDA and the GRIMM. Evaporation was then further investigated by measuring dried particles and reverting to the size distribution at the source. The SMPS was needed to measure small particle sizes, but measurements were no longer affected by evaporation as the particles were completely dried. Complete evaporation of the particles enables a better risk assessment in the context of airborne transmission. This implies that for specific applications, measurement instruments must be chosen carefully and particle drying always needs to be considered. Additional experiments must be carried out to clarify how measurements from the studied instruments can be achieved for particles that are not spherical.

References

1. Horrocks, M. W. H. Experiments made to determine the conditions under which specific bacteria derived from sewage may be present in the air ventilating pipes, drains, inspection chambers, ans sewers. Public Heal. 495–506 (1906).


**Acknowledgements**

This research was supported by a NSERC Alliance Grant (ALLRP545363/2019), Polytechnique Montréal. Xavier Lefebvre is supported by the Hydro-Québec excellence scholarship as well.

**Author contributions statement**

X.L. conceived the experiments, X.L. and A.S. conducted the experiment(s), X.L. analysed the results. All authors reviewed the manuscript.

**Additional information**

**Data Availability**

Data that support the findings of this study are available from the corresponding author upon reasonable request.

**Competing Interests**

The authors declare no competing interests.
Figure Legends and Tables

- **Figure 1.** Comparison of the four instruments with DEHS aerosols. Results are averaged over three measurements. Shaded areas show the standard deviation within repeated runs.

- **Figure 2.** Sampling losses observed with the GRIMM compared to the other three instruments. Results are averaged over three measurements. Shaded areas show the standard deviation within repeated runs.

- **Figure 3.** Comparison of the four instruments with water-based aerosols. Results are averaged over a triplicate of the measurements. Shaded areas show the standard deviation within repeated runs.

- **Figure 4.** Comparison of the GRIMM and the PDA to show the effect of the sampling tube on the measurements. Results are averaged over triplicate measurements. Shaded areas show the standard deviation within repeated runs.

- **Figure 5.** Size distribution from the SMPS with dried NaCl aerosols and the calculated size distribution at the source. Results are averaged over triplicate measurements. Shaded areas show the standard deviation within repeated runs.

- **Figure 6.** Schematic representation of the experimental setup
<table>
<thead>
<tr>
<th>Instrument</th>
<th>Size Range (µm)</th>
<th>Measurement time (s)</th>
<th>Sampling volume rate (L/min)</th>
<th>Advantages</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>PDA</td>
<td>0.5 - 8000</td>
<td>300</td>
<td>N/A</td>
<td>- No sampling line</td>
<td>- Calculated number concentrations</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- Large measurement range</td>
<td>- Loss of precision under 0.5 µm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- Measurement considered punctual</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- High particle detection resolution</td>
<td></td>
</tr>
<tr>
<td>GRIMM</td>
<td>0.25 - 32</td>
<td>6</td>
<td>1.2</td>
<td>- Portable</td>
<td>- Losses in sampling line</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- High measurement frequency</td>
<td>- Inadequate for wet aerosols</td>
</tr>
<tr>
<td>SMPS</td>
<td>0.022 - 0.671</td>
<td>76</td>
<td>1.2</td>
<td>- Goldstandard for submicron particles</td>
<td>- Limited measurement size range</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- High measurement frequency</td>
<td></td>
</tr>
<tr>
<td>Cascade Impactor</td>
<td>0.16 - 10</td>
<td>1800</td>
<td>10</td>
<td>- Portable</td>
<td>- Mass-based measurement</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- Allows for microbial analysis</td>
<td>- Losses in sampling line</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>- Limited size distribution resolution</td>
</tr>
</tbody>
</table>

**Table 1.** Comparison of measurement size range, measurement time, sampling volume rate, advantages and limitations of the size distribution measurement instruments.
<table>
<thead>
<tr>
<th><strong>Receiver focal length</strong></th>
<th>300 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Scattering angle</strong></td>
<td>30 deg</td>
</tr>
<tr>
<td><strong>Aperture mask</strong></td>
<td>Mask A for DEHS and Mask B for water-based</td>
</tr>
<tr>
<td><strong>Particle refractive index</strong></td>
<td>1.454 for DEHS and 1.334 for water</td>
</tr>
</tbody>
</table>

**Table 2.** Settings used on the PDA system