https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques Discussions

Aqueous particle generation with a 3D printed nebulizer

Michael Rösch[1,2] , and Daniel J. Cziczo[1,3,4]

[1]Department of Earth, Atmospheric & Planetary Sciences, Massachusetts Institute of Technology, Cambridge, 02139, USA

[2]Department of Environmental Systems Science, Eidgenössische Technische Hochschule - ETH, Zurich, 8092, Switzerland

[3]Department of Civil Environmental Engineering, Massachusetts Institute of Technology, Cambridge, 02139, USA

[4]Department of Earth, Atmospheric and Planetary Sciences, Purdue University, West Lafayette, IN 47907 USA

Correspondence to: Michael Roesch (michael.roesch@env.ethz.ch)

Abstract. In this study, we describe the design and testing of a high output stability constant liquid feed nebulizer using the Venturi principle to generate liquid particles from solutions. This atomizer, the PRinted drOpleT Generator (PROTeGE) was manufactured using stereolithography (SLA) printing. Different concentrations of ammonium sulfate solutions were used to characterize the size and number concentration of the generated particles. A comparison of a 3D printed 0.5 mm orifice with a more dimensionally accurate and symmetric machined 0.5 mm brass orifice using the same ammonium sulfate solutions was also performed. PROTeGE is also shown to be capable of dispersing polystyrene latex spheres (PSLs) for calibration purposes. The particle number concentrations obtained in this study ranged from ~10000 cm^{-3} for 0.75 µm to ~100 cm^{-3} for 5.0 µm PSL particles with a dependence on the concentration of the dispersed solution. PROTeGE is easy to manufacture and operate, low in maintenance, and cost-effective for laboratory and field generation of particles from aqueous media.

1 Introduction

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques Discussions

Reliable and cost-effective particle generation methods are necessary for applications where a well-defined mono- or polydisperse particle concentrations and size is required. High concentrations of monodisperse aerosol particles (>10^6 particles/cm^3) in the nm to μm diameter size range can be created by instruments utilizing vapor condensation and electrospray techniques. Similar concentrations and size ranges of aerosol particles can be produced from bulk solutions using a variety of instruments such as vibrating-orifice aerosol generators and ultrasonic nebulizers. Another common technique used to generate liquid aerosol particles is by pressurized-air nebulization. In this method, compressed air is utilized to shatter a solution into small aerosol droplets with a specific size distribution (Swiderska-Kowalczyk et al., 1997).

Pressurized-air nebulizers have been used in numerous studies which require the generation of aqueous aerosol particles. For example, Wang et al. (2019) used a nebulizer to create ammonium sulfate seed particles in cloud droplet activation studies. Kong et al. (2018) created aqueous aerosol particles used to study deliquescence and ice nucleation in sea salt particles. These are two of many studies focused on the influence of aerosol particles in the Earth's atmosphere. Pressurized-air nebulizers are also used in pharmaceutical applications to produce nanometer-sized drug particles with a specific size distribution (Eerikainen et al., 2003). It is clear that the pressurized-air nebulizer is a ubiquitous instrument for studies or applications requiring aqueous aerosol particle generation. Although effective, commercial particle generation instruments may not be economically feasible for all research and teaching institutions wishing to perform these types of experiments.

Advances in 3D printing have made it possible to rapidly fabricate high-resolution (μm-scale) devices. Stereolithography (SLA), a form of 3D printing technology, creates objects in layers through the use of photopolymerization. In conjunction with Computer Aided Design (CAD), or Computer Aided Manufacturing (CAM) software, an ultraviolet (UV) laser is used to trace a pre-programmed design on to the surface of a photopolymer contained in a vat. The resin is photochemically solidified and forms a single layer of the desired object. The Form 2 SLA 3D printer (Formlabs, Inc.), used in this work, is capable of creating objects with a layer thickness of 25 μm.

The Form 2 was previously used to fabricate PRIZE, a compact fluidized bed aerosol generator (Roesch et al. 2017). It was found that PRIZE was able to successfully disperse aerosol particles from dry

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques Discussions

material without creating artifact particles (particles generated from the material used to fabricate the generator). The impetus for this study, similar to the study presented in Roesch et al. 2017, was to fabricate a low-cost, constant pressure nebulizer, using a SLA 3D printer, PROTeGE.

2 Methods

2.1. Design

PROTeGE was designed using a computer aided design (CAD) program (Solidworks 2015, Dassault Systems). There are two versions: the first is printed as a single part including a 0.5 mm diameter orifice (Fig. 1), while the second, featuring the same inner and outer dimensions, has an exchangeable nozzle to use various machined orifice diameters. The orifice used in this study is within a commercially available brass nozzle (Part Number 6183T63, McMaster-Carr) that is screwed into the pressurized air inlet of PROTeGE (Fig. 1d). This modular feature enables rapid exchange of nozzles with a different orifice diameter using the same printed unit. In contrast, the exclusively printed version of PROTeGE has a fixed orifice diameter for continuous and simple operation. Both versions are based on the generator designs of May, 1973 and Liu & Lee, 1975. Both versions include a printed GL45 bottle cap so that the aqueous material bottle can to directly attached to the nebulizer body. Unlike the repurposing of existing nebulizers built for other uses, such as medical applications (Reisner et al., 2001), PROTeGE was designed specifically for research applications e.g. instrument calibration and particle generation, similar to other custom-built nebulizers (Wex et al., 2015). In contrast to other generation systems, which are typically machined, PROTeGE is made from photopolymer resin (e.g. FLGPCL02, Formlabs Inc.), weights only ~50 g, and can therefore feature smaller overall dimensions.

The front inlet to PROTeGE is a 6.35 mm (0.25 in) barbed tube to connect to a pressurized airflow. The inlet is end-capped by the orifice. Directly following, and perpendicular oriented to the orifice, is the liquid feed 3.18 mm (0.125 in) inlet to the dispersible solution. Excess liquid from the nebulizing process exit the chamber through a 6.35 mm outlet at the bottom, dripping directly back into the feed bottle. Aqueous particles exit the chamber through the 9.53 mm (0.375 in) aerosol outlet at the top. The overall dimensions of PROTeGE are 17 x 45 x 65 mm (width, depth, height). Designed CAD files were converted to style files (.stl) to be readable by the 3D printer software (PreForm, Formlabs Inc.).

2.2. Manufacturing

The manufacturing and post-processing of the parts was performed as described by Roesch et al., 2017 using the same 3D printer software, clear photopolymer resin (FLGPCL02, Formlabs Inc.) and 3D SLA printer (Form 2, Formlabs Inc.). Modifications were made to the dimensions of the default contact points of the printing scaffolding; in this study scaffolding was reduced to 0.45 mm due to the overall smaller geometry of PROTeGE (i.e., lower mass needing to be supported). Using a resolution of 100 µm, eight complete PROTeGEs can be printed on the build surface at the same time, taking ~8 h.

A custom UV box was used to post-cure the printed parts. Inside the box, the printed parts were placed on a slow moving turntable to be illuminated equally from all sides by 28 high-power LEDs emitting at 405 nm. It should be noted that the curing time depends on the size and wall thickness of the printed part; one hour per mm wall thickness is suggested. The post-curing time for PROTeGE was ~1 h. For more detailed information see the manufacturing section in Roesch et al., 2017.

2.3. Experimental setup

A schematic of the experimental setup including the relevant flow rates used in this study is shown in Fig. 2. Dry, filtered, pressurized air was used as the carrier gas. The input flow rate of 1.7 L min^{-1} (at 35 psi) into PROTeGE was controlled by a rotameter (MR3A, Omega Engineering). A high velocity jet is created by the expansion of the pressurized air through the orifice. As a result, the pressure behind the orifice drops, the liquid is pulled upward from the feed bottle, and the high velocity jet disperses the liquid solution into droplets. Large droplets that are unable to follow the streamlines through the aerosol outlet are removed by impaction at the curved wall; these drip back as excess liquid into the feed bottle through a drain outlet at the bottom of PROTeGE. For these experiments, the droplets from the aerosol outlet were subsequently dried using a silica gel drier. Downstream, the flow of residuals (remaining solid cores of the droplets) was split into two flows. The first is sent to an optical particle sizer (OPS, Model 3330, TSI Inc.) to determine particle number size distributions (PNSD) in the size range of 0.3 to 10 µm and the remainder through a filter (IDN-4G, Parker) open to lab. Unless otherwise noted, all experiments

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques Discussions

presented here were performed using the brass nozzle with a 0.3 mm orifice and the previously stated pressures and flow rates.

3. Results

In this study three types of experiments were conducted to demonstrate the performance of PROTeGE: (1) An aerosol production experiment using four different sizes of polystyrene latex spheres (PSL's, Polysciences Inc., NIST traceable) ranging from 0.75 to 5.0 µm. (2) An application experiment where different concentrated ammonium sulfate solutions were dispersed and monitored over time. (3) An experiment comparing the performance of a printed 0.5 mm orifice to a 0.5 mm brass orifice using an ammonium sulfate solution. For all experiments the PNSD of the droplet residuals (i.e., after drying) was measured with the OPS.

Prior to each experiment, PROTeGE was immersed in a jar with double distilled deionized (DDI) 18.2 MΩ*cm Millipore water and sonicated for 10 minutes in an ultrasonic bath to ensure clean inner surfaces. Afterwards, PROTeGE was dried using pressurized nitrogen and connected to the setup. Each of the four samples was prepared in a separate 100 ml glass bottle, using 80 ml of DDI water plus multiple drops of the respective PSL solution (0.75 µm, 1.5 µm, 2.0 µm, 5.0 µm). The generated number concentration of PSL particles strongly depends on the concentration of the prepared PSL sample. Therefore, the higher the concentration of the solution, the higher the generated particle number concentration. A time-series measurement was performed for each of the four PSL samples. The obtained PNSD showed particle number concentrations from ~10000 cm^{-3} for 0.75 µm PSL particles to ~100 cm^{-3} for 5.0 µm PSL particles (Fig. 3). All four investigated PSL samples showed narrow PNSD's except the 5.0 µm PSL sample where a fraction of sub-micrometer particles was detected (Fig. 3d). This fraction of particles likely originates from the solution matrix the PSLs are suspended in. These data show that the curved design of the chamber makes PROTeGE capable of dispersing PSL particles with diameter up to at least 5.0 micrometers.

In addition to the PSL measurements, ammonium sulfate experiments with different solution concentrations (0.1 g L^{-1}, 0.6 g L^{-1}, 5.0 g L^{-1}) and an experiment to determine the performance of the printed 0.5 mm orifice versus the 0.5 mm brass orifice using the same ammonium sulfate solution of 0.6

Atmospheric Measurement Techniques Discussions

g L^{-1} were conducted. The three solutions were again prepared in separate 100 ml glass bottles using DDI water. The cleaning procedure of PROTeGE was identical to the one described above for the PSL experiments.

For the lowest concentration of aqueous ammonium sulfate solution (0.1 g L^{-1}) the maximum particle number concentration of ~14000 cm^{-3} was observed in the 0.3 µm bin of the OPS (Fig. 4a). The overall width of the generated PNSDs ranged from 0.3 µm up to 0.6 µm. Compared to the higher concentrated solutions of 0.6 g L^{-1} and 5.0 g L^{-1} this is rather narrow. Dispersing the 0.6 g L^{-1} solution of ammonium sulfate generated PNSDs in the range of 0.3 µm up to 1.5 µm with a maximum particle number concentration of ~8200 cm^{-3} in the 0.5 µm detection bin (Fig. 4b). For the highest concentration of 5.0 g L^{-1} the generated PNSDs ranged from 1.0 µm to 2.4 µm with a maximum particle number concentration of ~7600 cm^{-3} (Fig. 4c).

Overall the generated particle number concentrations for the different tested ammonium sulfate solutions will be sufficient enough to operate particle size selection instruments downstream of PROTeGE, e.g. a differential mobility analyzer (DMA), assuming ~10% of the introduced particles are selected as monodisperse aerosol particles.

In order to investigate the performance of an integrally printed 0.5 mm orifice versus the commercial brass nozzle the same 0.6 g L^{-1} ammonium sulfate solution was used. Visual observations of the printed 0.5 mm orifice showed that there were sometimes imperfections and asymmetries in the roundness of the printed orifice. It was therefore necessary to post-drill the printed orifice with a 0.5 mm bit by hand, and this was done for the PROTeGE used in experiments described here. The generated mean particle number concentration with the printed orifice was ~4500 cm^{-3} versus ~2400 cm^{-3} for the brass nozzle. Both orifices generated similar PNSDs with their maximum particle number concentrations at 0.3 µm leveling out to 1.0 µm at largest (Fig. 5).

4. Conclusion

In this study, we described the design and the performance of a low-cost particle generator – PROTeGE. The experiments presented here show the PROTeGE capability for generating four different sizes of PSL particles between 0.75 and 5.0 µm with resulting particle number concentrations between 100 cm^{-3} and

~10000 cm^{-3}. This enables PROTeGE to be used as a simple and cost effective particle generation unit for calibration purposes. In addition, we show the results of experiments using different concentrations of ammonium sulfate solutions. The generated PNSDs ranged from 0.3 µm up to 2.4 µm with stable output concentrations demonstrating that particle size selection instruments can be used downstream of PROTeGE. Finally, we compared the performance of a commercially machined brass 0.5 mm orifice to a 3D printed 0.5 mm orifice. Both orifices performed well, while the mean particle number concentration generated with the printed orifice was almost two times higher than the particle number concentration generated with the brass orifice. Due to the low cost of PROTeGE multiple generators can be used in parallel to reduce experimental time while running more samples.

Data availability: The .stl files for PROTeGE are available up on request.

Authors contribution: MR and DJC contributed both equally to the manuscript. The experiments were conducted by MR.

Competing interests: The authors declare that they have no conflict of interest.

References

Eerikainen, H., Watanabe, W., Kauppinen, E. I. and Ahonen, P. P: Aerosol flow reactor method for synthesis of drug nanoparticles, Euro. J. Pharma. and Biopharma., 55,3, 357-360, 2003.

Kong, X., Wolf, M., Roesch, M., Thomson, E. S., Bartles-Rausch, T., Alpert, P. A., Ammann, M., Prisle, N. L., and Cziczo, D. J.: A continuous flow diffusion chamber study of sea salt particles acting as cloud nuclei: deliquescence and ice nucleation, Tellus B, 70, doi:10.1080/16000889.2018.1463806, 2018.

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Liu, B. Y. H., and Lee, : An Aerosol Generator of High Stability, Am. Ind. Hyg. Assoc. J., 36, 861–865, 1975.

May, K.R.: The Collusion Nebulizer: Description, Performance and Application, J. Aero. Sci., 4, 235–243, 1973.

Reisner, C., Katial, R. K., Bartelson, B. B., Buchmeir, A., Rosenwasser, L. J., Nelson, H. S.: Characterization of aerosol output from various nebulizer/compressor combinations, A. All. Asth. Immun. , 1081-1206, 86, 5, 566-574, 2001.

Roesch, M., Roesch, C., and Cziczo, D. J.: Dry particle generation with a 3-D printed fluidized bed generator, Atmos. Meas. Tech., 10, 1999–2007, 2017.

Swiderska-Kowalczyk, M., Gomez, F. J., Martin, M.: Particle generation methods applied in large-scale experiments on aerosol behavior and source term studies, Inform. Tech. Cie., 819, 1997.

Wex, H., Augustin-Bauditz, S., Boose, Y., Budke, C., Curtius, J., Diehl, K., Dreyer, A., Frank, F., Hartmann, S., Hiranuma, N., Jantsch, E., Kanji, Z. A., Kiselev, A., Koop, T., Möhler, O., Niedermeier, D., Nillius, B., Rösch, M., Rose, D., Schmidt, C., Steinke, I., and Stratmann, F.: Intercomparing different devices for the investigation of ice nucleating particles using Snomax® as test substance, Atmos. Chem. Phys., 15, 1463–1485, 2015.

Wang, J., Shilling, J. E., Liu, J., Zelenyuk, A., Bell, D. M., Petters, M. D., Thalman, R., Mei, F., Zaveri, R. and Zheng, G.: Cloud droplet activation of secondary organic aerosol is mainly controlled by molecular weight, not water solubility, Atmos. Chem. Phys. 19, 941-954, 2019.

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

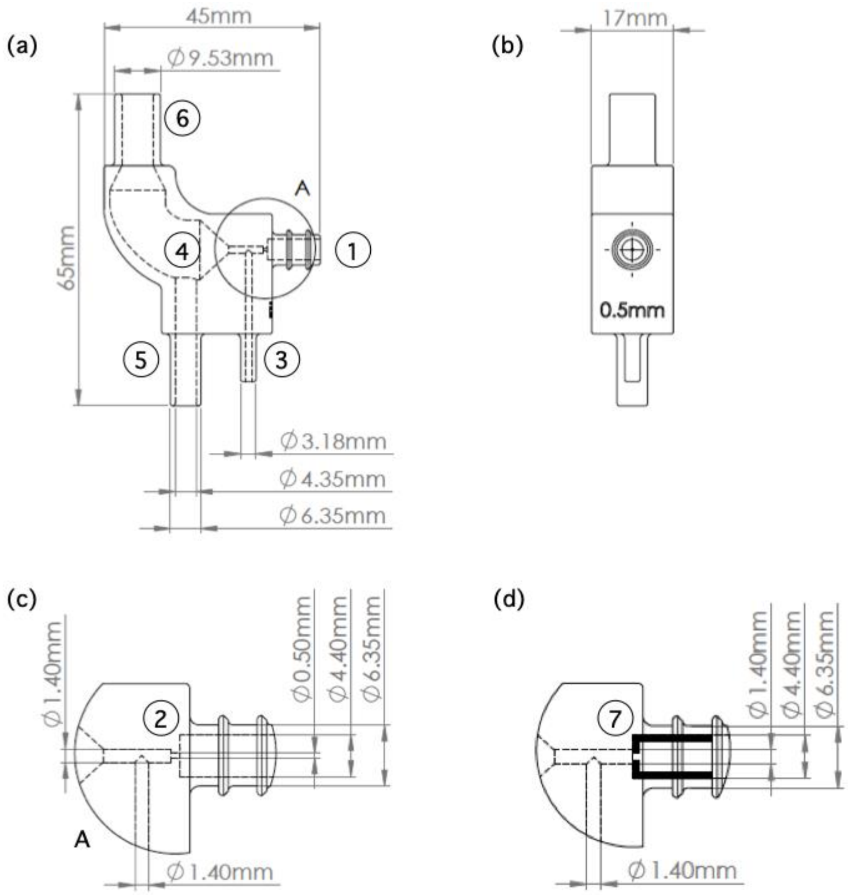

Figure 1: Dimensioned drawings of PROTeGE: (a) side view; (b) front view; (c) detailed view of the inlet section with the printed 0.5mm orifice; (d) detailed view of the inlet section with the exchangeable nozzle/orifice. PROTeGE consists of: (1) a pressurized air inlet, (2) a printed 0.5mm orifice, (3) a liquid feed inlet, (4) a central impaction chamber, (5) the drain outlet, (6) an aerosol outlet, and an optional (7) exchangeable orifice.

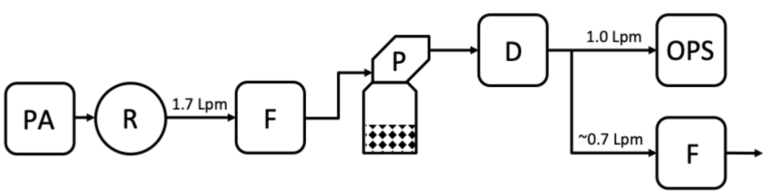

Figure 2: Schematic of the experimental setup used in this study. A dry pressurized air flow (PA) was passed through a rotameter (R) to control the flow rate and a filter (F) upstream of PROTeGE (P). Generated droplets were dried with silica gel (D) before the flow of particles was directed into an OPS with excess flow discarded through a filter (F).

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques
Discussions

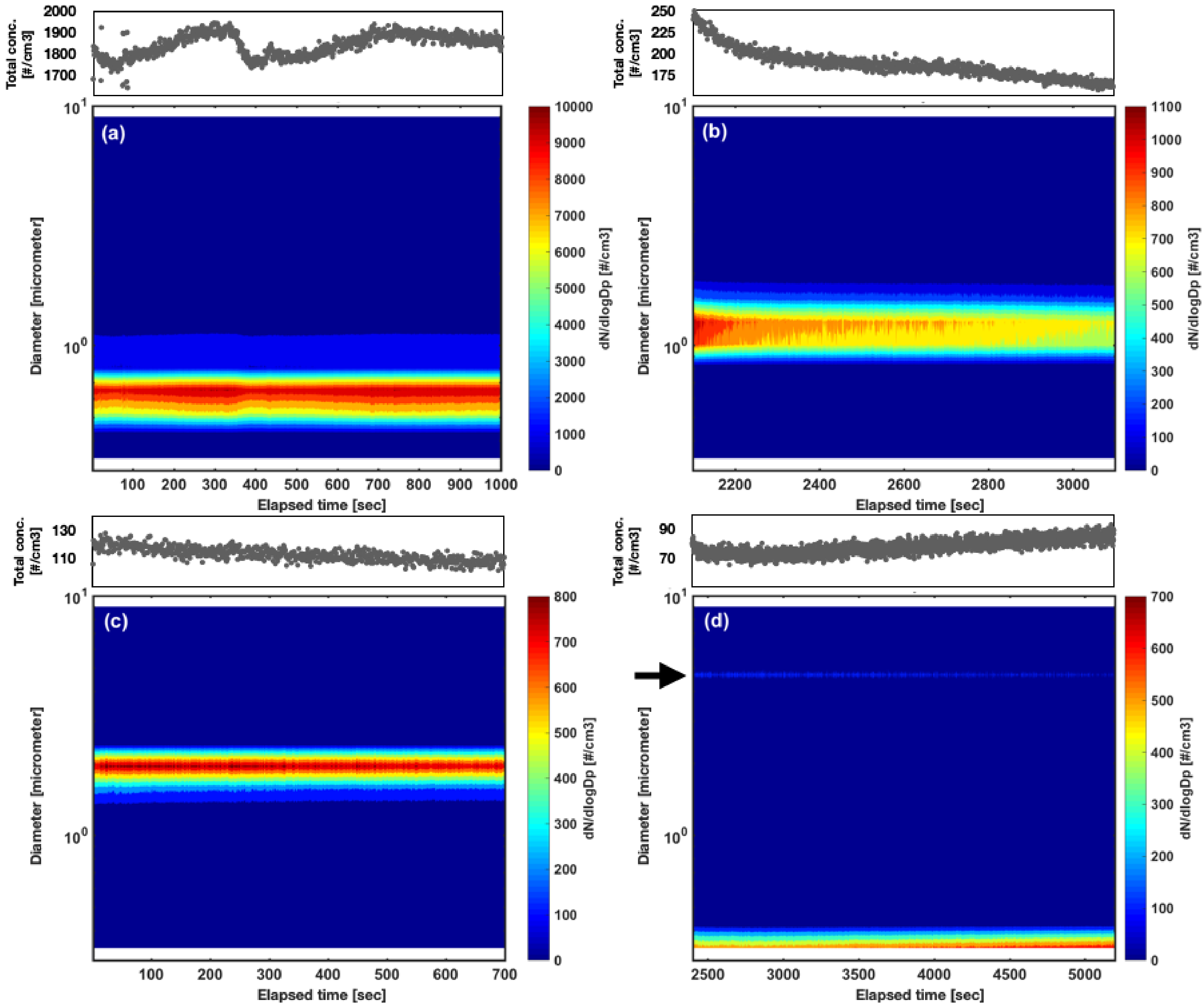

Figure 3: Total particle number concentrations and particle number size distributions as function of time for different sized polystyrene latex spheres generated with PROTeGE and detected by the OPS: (**a**) 0.75 µm; (**b**) 1.5 µm; (**c**) 2.0 µm; (**d**) 5.0 µm where the black arrow denotes the PSL size and the small particles represent atomized matrix material (see text for details).

https://doi.org/10.5194/amt-2020-155
Preprint. Discussion started: 9 June 2020

Atmospheric Measurement Techniques
Discussions

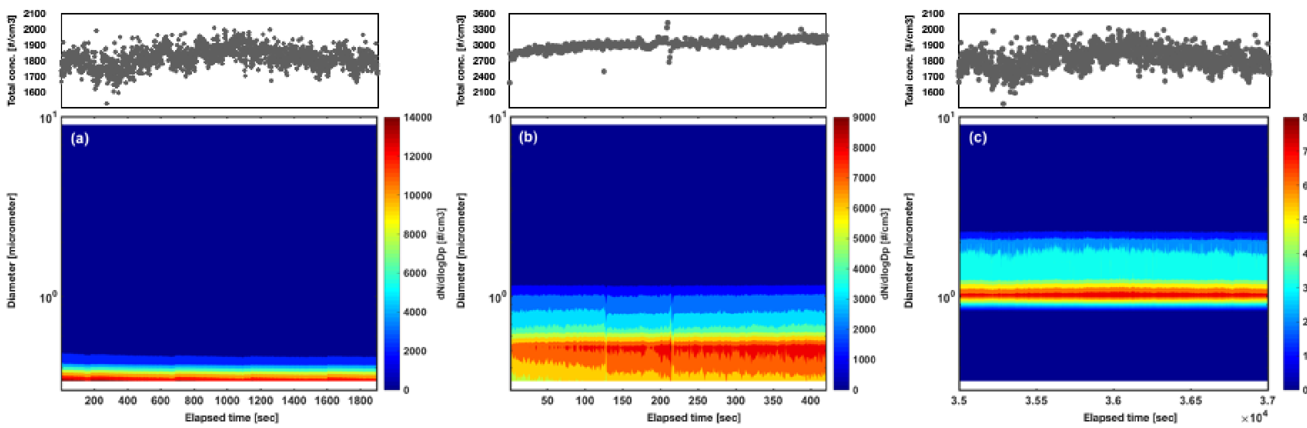

Figure 4: Total particle number concentrations and particle number size distributions as a function of time and of concentration of aqueous ammonium sulfate: **(a)** 0.1 g L^{-1}; **(b)** 0.6 g L^{-1}; **(c)** 5.0 g L^{-1}.

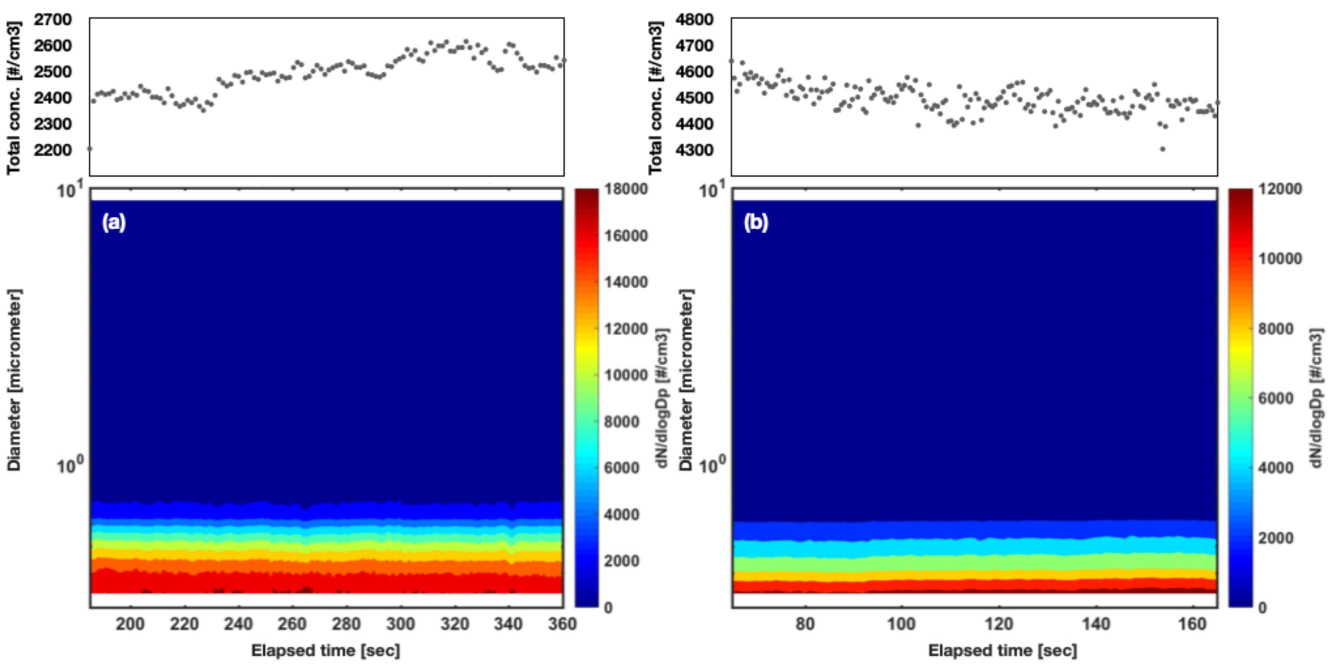

Figure 5: Total particle number concentrations and particle number size distributions of aqueous ammonium as a function of time for sulfate particles at 0.6 g L^{-1}: **(a)** 0.5mm printed orifice; **(b)** 0.5mm brass orifice.