Technical Note

**A Sampler for Collecting Fine Particles into Liquid Suspensions**

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**ABSTRACT**

In this study, we developed a particle sampler for aerosol suspensions (PSAS) that collects ambient PM₂.₅ directly as liquid suspensions. The PSAS, which operates at a sampling rate of 50 L min⁻¹, first pre-selects fine particles with a custom-designed PM₂.₅ inlet and then enlarges them into 2–5 µm droplets via condensational growth with a saturation-condensation unit. Subsequently, the enlarged droplets are sequentially collected in our novel collection impactor as liquid suspensions. Laboratory evaluations indicate good collection efficiencies for the PSAS with both polydisperse and monodisperse fine particles, with 85% of the particles being effectively preserved in the collected suspensions. Field evaluations suggest good agreement between the PSAS suspensions and parallel filter samples for the analyzed chemical components (water-soluble Fe(II) and Cr(VI)) in the ambient PM₂.₅. Thus, the PSAS is suitable for long-term particle collection and can be used to facilitate the online chemical and toxicological analysis of ambient PM₂.₅.

**Keywords:** Aerosol sampler; PM₂.₅; Suspension samples; Condensational growth; Droplet impactor.

**INTRODUCTION**

Adverse health outcomes caused by exposure to particulate matter (PM), especially PM₂.₅ (particles with aerodynamic diameter equal or less than 2.5 µm), have been well documented (Lelieveld et al., 2015; Zhao et al., 2018). The PM-induced health effects are strongly depending on the chemical and toxicological properties of particles (Charrier and Anastasio, 2012; Saffari et al., 2015). Conventional PM analysis usually involves filtration or inertial impaction processes in which particles are collected on substrates such as filters or foils, and PM chemical and toxicological analysis are performed offline. Although these methods have been widely adopted, they are associated with drawbacks, including long time intervals for sampling and intensive labor for sample preparation and analysis, underestimations of insoluble PM components due to incomplete extractions (Wang et al., 2013b; Gao et al., 2017), as well as potential sampling artifacts for volatile species during long-term sampling (Eatough et al., 2003). Efforts have been made to eliminate particle collection procedure, and to accomplish real-time direct measurements/analysis of particle chemical composition by introducing advanced mass spectrometers (Jayne et al., 2000; Smith et al., 2004; Ng et al., 2011). These techniques have significantly contributed to the further understanding of PM chemical characteristics, as well as their potential sources and formation mechanism (Canagaratna et al., 2007; Smith et al., 2010; Sun et al., 2012).

Another alternative is to capture particles into liquid as suspension samples which can be directly analyzed. Such techniques include applying wet collection surfaces into filtration or impaction devices (Cofer et al., 1985; Buhr et al., 1995; Phan and McFarland, 2004; Takeuchi et al., 2005; Orsini et al., 2008; Puthussery et al., 2018), as well as devices involving condensational growth method (Okuyama et al., 2007; Dong et al., 2012). In later techniques, the sampled particles are firstly mixed with vapors of working fluids (i.e., water, butanol etc.) (Hering and Stolzenburg, 2005). The mixture of particles and vapors is then cooled down to promote vapors condensing on particle surfaces, and enlarge particles into larger droplets that are easier to be detected (i.e., with a condensation particle counter (CPC)) (Stolzenburg and Mcmurry, 1991; Hering and Stolzenburg, 2005; Hering et al., 2014), or collected directly as suspension samples. For example, a steam jet aerosol collector (SJAC) was developed by Khlystov et al. (1995) and was further enhanced by Slanina et al. (2001), in which particles are mixed with water steam for condensational growth and the grown droplets are...
then collected by cyclones. Another widely used technique is the Particle-into-Liquid Sampler (PILS) (Weber et al., 2001; Orsini et al., 2003). In PILS, the grown droplets are collected by a single-nozzle impactor, and the liquid sample collected on the impactor surface is removed by a small, constant flow of purified water that also contains an internal standard. Both SJAC and PILS have been adopted for online measurements of ionic components of PM in combination of ion chromatograph (IC) (Perrino et al., 2015; Stieger et al., 2019). A similar aerosol sampler was introduced by Wubulihairen et al. (2015a, b), in which a condensed water splitter is included to prevent sample dilution. Kidwell and Ondov (2001) also reported a high-flow-rate sampler with flow rate up to 170 L min⁻¹. These advanced techniques have effectively overcome some drawbacks in conventional particle collection methods.

There have been concerns on the change of chemical properties of sampled particles due to the usage of high-temperature water steam, as well as the decrease of pollutants’ concentrations in liquid suspensions due to the usage of transport flow on collection surface (Perrino et al., 2015). As a different approach, the Versatile Aerosol Concentration Enrichment System (VACES) in tandem with a liquid impinger (BioSampler; SKC-West Inc., Fullerton, CA, USA) was developed to collect PM directly as aqueous suspension samples (Kim et al., 2001a, b). In VACES-BioSampler tandem, a water tank is used as saturator where water is gently warmed up to generate vapors, instead of using high-temperature steam generation devices. Based on the condensational growth components of VACES, Wang et al. (2013b, 2016) introduced a high-flow-rate (200 L min⁻¹) aerosol-into-liquid collector in which grown droplets are directly collected and accumulated as liquid suspensions without introducing transport flow onto collection surface, and further deployed this PM collector as online PM metal monitors. Recently a new water-based condensational growth technology was reported, in which a tandem of “conditioner-initiator-moderator” is included in order to improve condensational growth performance (Hering et al., 2014). It has been applied into a liquid spot sampler to collect particles as suspensions (Fernandez et al., 2014). These devices have provided new insights on the developments of advanced particle collection techniques for online analysis of chemical and toxicological properties of PM.

In order to achieve relatively high pollutant concentrations in collected liquid suspensions and good preservation of their chemical properties, a relatively high sampling flow rate, a gentle generation of water vapor, as well as exemption of the transport flow on collection surface are favorable for advanced PM collection techniques. Here we present a particle sampler for aerosol suspensions (PSAS) for collecting fine particles directly as liquid suspension samples with a flow rate of 50 L min⁻¹. The PSAS contains a custom-designed PM₂.₅ inlet, a saturator tank and a condenser for condensational growth, as well as a novel single-nozzle collection impactor for collecting the grown droplets as suspension samples. The collection impactor utilizes several unique configurations compared to conventional inertial impactors, including a Teflon gasket for restraining impacted droplets from bouncing back to air stream, and a mesh surrounding the impaction surface to wick the collected droplets into sample outlet. Both laboratory and field evaluations indicate that the PSAS is capable for ambient PM collections and can be further modified for online monitoring of PM chemical and toxicological characteristics.

METHODS

Description of the System Configuration and its Components

The schematic of the PSAS system configuration is presented in Fig. 1. The PSAS has a design flow rate of 50 L min⁻¹ and contains four major components: the PM₂.₅ inlet, the saturator, the condenser and the collection impactor. The PM₂.₅ inlet is a custom-designed impactor containing two slit nozzles (each nozzle is 1 mm in width and 57.6 mm in length) which is designed to have a 50% cutpoint of 2.5 µm under a flow rate of 50 L min⁻¹. While for the collection impactor of PSAS, it is a single-round-nozzle impactor (nozzle diameter: 4 mm) designed to have a 50% cutpoint of 1.5 µm under a flow rate of 50 L min⁻¹. For inertial impactors, the 50% cutpoint size is determined by Stokes number as below (Hinds, 1999):

\[
Stk = \frac{\rho_p d_p^3 u_C}{9 \eta D_j}
\]

where \(Stk\) is the Stokes number, and in current designs 0.59 and 0.24 were used for rectangular slit nozzle and round nozzle, respectively (Hinds, 1999). \(\rho_p\) is the particle density (1000 kg m⁻³), \(d_p\) represents the diameter of a particle having a 50% probability of impaction. \(u\) is the jet velocity at the nozzle, which is calculated based on designed flow rate and nozzle dimension. \(C\) is the Cunningham slip correction factor, and \(\eta\) is the dynamic viscosity of the air (1.8 × 10⁻⁵ kg m⁻¹ s⁻¹). \(D_j\) is the geometry dimension of nozzle(s) in the impactor, which is the width for rectangular nozzle and the diameter for round nozzle.

In the PSAS system, the sampled air firstly passes through a PM₂.₅ inlet for preliminary size selection. PM₂.₅ then enters the saturator (stainless steel, 400 mm × 150 mm × 120 mm in dimension) filled with deionized water for saturation, in which a heater is deployed to gently heat up the water. The temperature of aerosol-water vapor mixture (measured by a type K thermal couple) is maintained at around 38°C by adjusting the voltage applied to the heater. The saturated mixture then passes the condenser (500 mm in length and 20 mm for inner diameter) which is connected to a circulating chiller. The coolant in the chiller is maintained at 4–5°C to cool down the mixture to 21–22°C, promoting particle condensational growth and finally enlarging particles into droplets with larger sizes. The condenser is slightly angled from horizontal in order to assist the drainage of condensed water accumulating on the tube inner surface. The grown droplets are finally collected via inertial impaction in the collection impactor. The new designed collection impactor utilizes several unique configurations. Different from
Fig. 1. System schematic of particle sampler for aerosol suspensions (PSAS). The inserted figure shows the structure of the collection impactor.

conventional impactors, a Teflon gasket is placed above the quartz impaction surface in order to create a groove surrounding the impaction surface. Such a groove serves as the droplets’ accumulation/transportation area in combination of a stainless steel mesh around the impaction surface’s outside edge as a wicking material (Orsini et al., 2003). When droplets are impacted on the quartz surface, the spreading airflow blows the impacted droplets into the groove, but the air streams turn around at the inner surface of the Teflon gasket. Therefore, impacted droplets are restrained from bouncing back into airflow, resulting in better collection efficiencies. A multi-channel peristaltic pump continuously transfers condensed water from the condenser and the collection impactor, as well as the collected droplets. Although PSAS has a medium sampling flow rate (i.e., 50 L min⁻¹) compared to some previously reported samplers with very high sampling flow rates (Kidwell and Ondov, 2001; Kim et al., 2001a, b; Wang et al., 2013a), it is more flexible to move and to be deployed into field collections due to its smaller size and lighter weight. All components and accessories of PSAS are mounted on a custom-built wheeled aluminum frame, with a dimension of 1000 mm × 700 mm × 800 mm and a total weight of 40 kg.

Laboratory Evaluations of PSAS Collection Efficiency

The first part of laboratory evaluations of PSAS is to characterize the collection efficiencies of the PM₂.₅ inlet and the collection impactor. Polydisperse NaCl particles, with a peak diameter around 3–3.5 µm, were generated by a supermicron particle generation system (Chen et al., 2016). Briefly, NaCl solution was injected via a syringe pump into an ultrasonic nozzle (60 KHz, Model 8700-60; Sono-Tek Corp., Milton, NY, USA) motored by an ultrasonic generator (Model 06-05018; Sono-Tek Corp., Milton, NY, USA) to generate micron-sized NaCl droplets. A mixing chamber was placed beneath the ultrasonic nozzle where filtered air was supplied and mixed with generated droplets for dilution and stabilization. The tested units (i.e., the PM₂.₅ inlet and the collection impactor) were placed inside the chamber and particle size distributions before and after the tested units were measured by an Aerodynamic Particle Sizer (APS Model 3321; TSI Inc., Shoreview, MN, USA). The collection efficiencies of tested units were determined by comparing the upstream and downstream APS measurements.

In the second step of laboratory evaluations, the performance of condensational growth within PSAS under certain operation parameters was examined. Polydisperse ammonium fluorescein particles were generated by an atomizer and mixed with HEPA-filtered make-up air. Before entering PSAS, the size distributions of intake ammonium fluorescein particles were detected by a Scanning Mobility Particle Sizer spectrometer (SMPS), which contains an electrostatic classifier (Model 3081; TSI Inc., Shoreview, MN, USA), a soft X-ray neutralizer (Model 3088; TSI Inc., Shoreview, MN, USA), a Differential Mobility Analyzer (DMA Model 3081A; TSI Inc., Shoreview, MN, USA) and a condensation particle counter (Model 3772; TSI Inc., Shoreview, MN, USA). During the evaluation tests, the saturator and condenser of PSAS were maintained at 38 and 5°C, respectively. These temperature parameters were selected among different
temperature combinations who has the highest overall collection efficiencies of PSAS, and were consistently used in all evaluation tests throughout this manuscript. The size distribution of grown droplets after condensational growth was measured by APS at the exit of condenser. The measured size distributions for intake particles and grown droplets are normalized by their corresponding peak concentrations, since different types of instrument were used for intake particles and grown droplet measurements (i.e., SMPS for intake particles and APS for grown droplets).

Lastly, the overall collection efficiencies of PSAS were determined using polydisperse/monodisperse ammonium fluorescein particles generated by atomizer. It should be noted that ammonium fluorescein particles are highly hydrophilic, therefore they may not fully represent hydrophobic particles generated from certain sources (i.e., combustions). The experiment system schematic is shown in Fig. 2. For polydisperse tests, the generated particles were mixed with HEPA-filtered air and collected by PSAS. For monodisperse tests, the generated particles were classified by the neutralizer-DMA tandem mentioned previously, and a certain size of particles were selected and directed into PSAS after dilution. A portion of sampled particles were directed into a 5 L min\(^{-1}\) sampling line where a 47 mm quartz filter was placed to collect upstream sample. Another 47 mm quartz filter was placed at the exhaust flow of collection impactor to capture particles escaped from PSAS. After each collection, the upstream and downstream filters were extracted by 20 mL of 10% ammonia solution as upstream and downstream sample extracts. Besides, the condensed water from condenser was also preserved to evaluate particle depositions/losses within the condenser. The residuals on the nozzle and impaction surface of collection impactor were also extracted by 20 mL of 10% ammonia solution to evaluate the particle losses on nozzle and impaction surface. Extracts of upstream and downstream filters, condensed water in condenser, extractions of nozzle and impaction wall, as well as the actual suspension sample collected by PSAS were measured for their fluorescent signals. The fluorescent measurements were performed using a module fluorescent monitor, which contains a LED light source with wavelength of 470 nm (LLS-470; Ocean Optics, Inc., Dunedin, FL), a fluorescent flow cell (FIA-SMA-FL-ULT; Ocean Optics, Inc., Dunedin, FL), and a portable spectrometer (USB4000-UV-VIS; Ocean Optics, Inc., Dunedin, FL). Based on fluorescent signals from upstream filter extracts, percentage of particle depositions in condenser, collection impactor nozzle and impaction surface, as well as actual particle collection in suspension sample and particles escaped into exhaust were calculated based on corresponding fluorescent signals and flow rates (the unapportioned percentage is defined as “others”).

Field Evaluations of PSAS Performance and Comparison with Parallel Filters

After laboratory evaluations, the PSAS system was deployed in the field for collections of ambient PM\(_{2.5}\). The field sampling campaign were carried out at the 6\(^{th}\) floor of Sino-Italian Tsinghua Ecological and Energy Efficient Building in Tsinghua University during September and October in 2019. The building is located 100–200 m from a local street with some traffic densities during most of the day, as well as restaurants nearby (Ren et al., 2019). Ambient PM\(_{2.5}\) were collected as suspension samples via the PSAS system and each sample was collected for 3 hours. In parallel, PM\(_{2.5}\) were also collected by conventional filter sampling method, in which a PM\(_{2.5}\) cyclone (URG-2000-30EH; URG Corp., Chapel Hill, NC, USA) with flow rate of 16.7 L min\(^{-1}\) was used as sampling inlet followed by a 47 mm quartz filter for particle collection. In total 12 sets of samples (i.e., suspension and parallel filter) were collected. It should be noted that the main purpose of current field sampling campaign is to demonstrate the agreements for analyzed chemical components between suspensions collected by PSAS and conventional filter sampling, therefore the sampling hours were not specifically set and were randomly chosen during daytime.

![Fig. 2. Experimental setup for testing the collection efficiency.](image-url)
Concentrations of water-soluble Fe(II) and Cr(VI) in collected samples were determined by spectrophotometry, similar to the method reported in previous studies (Sowlat et al., 2016; Wang et al., 2016). Fe(II) and Cr(VI) were measured since they are important metal species participating/promoting the reactive oxygen species in human body (Valko et al., 2005; See et al., 2007). Briefly, the quartz filter samples were extracted in 20 mL of deionized water with assistance of sonication for 45 minutes. Both filter extracts and suspensions were filtered by 0.20 µm polypropylene syringe filters to separate the insoluble components. For Fe(II) measurements, ferrozine reagent was added into measured samples (5% v/v) and standard curves was determined by measuring Fe(II) standard solutions (0–100 ppb) prepared by serial dilution. For Cr(VI) measurements, diphenylcarbazide reagent was added into measured samples (5% v/v) and standard curves was determined by 0–20 ppb standards. Spectrophotometry measurements were performed using a deuterium and tungsten halogen light source (DT-MINI-2-GS; Ocean Optics, Inc., Dunedin, FL), a 100 cm Liquid Waveguide Capillary Flow Cell (LWCC 3100; WPI Inc., Sarasota, FL, USA) and a portable spectrometer (USB4000-UV-VIS; Ocean Optics, Inc., Dunedin, FL). Maximum absorptivities were observed and recorded at 562 and 540 nm for Fe(II) and Cr(VI) measurements, respectively. Finally, the measured concentrations were converted to corresponding ambient concentration based on sampling flow rate and time.

RESULT AND DISCUSSIONS

Characterization of PM$_{2.5}$ Inlet and Collection Impactor

Laboratory evaluations indicate good particle separation/collection performance for the PM$_{2.5}$ inlet and the collection impactor. The experimentally measured collection efficiency curves for the PM$_{2.5}$ inlet and the collection impactor of PSAS are presented in Fig. 3(a)–(b). It can be seen that the 50% cutpoint size is approximately 2.2 and 1.2 µm for PM$_{2.5}$ inlet and collection impactor, respectively. These experimentally determined 50% cutpoint sizes agree well with the theoretically designed values (2.5 and 1.5 µm for PM$_{2.5}$ inlet and collection impactor, respectively). In addition, collection efficiency curve of the PM$_{2.5}$ inlet is also compared to International Standard for PM$_{2.5}$ separation efficiency curve (ISO 7708) (ISO, 1995). As shown in Fig. 3(a), the collection efficiency curve of the designed PM$_{2.5}$ inlet has similar 50% cutpoint size as ISO 7708 standard curve, but a steeper slope over the size separation range compared to ISO 7708, indicating the good separation performance in the current designed PM$_{2.5}$ inlet.

Evaluations on Condensational Growth

In PSAS, we show that submicron particles can be effectively grown to larger droplets for sequential collection. The size distributions of intake particles and grown droplets after condensational growth are measured, which is presented in Fig. 4. As shown in Fig. 4, the sizes of intake particles are mostly located in submicron ranges, with a peak diameter around 120 nm. After condensational growth, the intake particles are enlarged into micron droplets, with a peak diameter between 2–5 µm. These grown droplets can be effectively collected by the following collection impactor, since majority of these particles are larger than the 50% cutpoint size of collection impactor as indicated in previous section. Moreover, the size distributions of grown droplets also validate the choice of temperature parameters (i.e., saturation and condensation temperatures of 38 and 5°C, respectively), under which the intake particles are grown to certain sizes above the cutpoint of collection impactor, but not oversized to cause significant losses at the impactor nozzle (shown in the following sections).

Evaluations of Overall System Collection Efficiency

Good overall system collection efficiencies were obtained for polydisperse (with particle size distribution similar to intake particles in Fig. 4) and DMA-selected monodisperse ammonium fluorescein particles. The collection efficiencies
Fig. 4. Normalized size distributions for intake particles and grown droplets. The measured size distributions are normalized by their corresponding peak concentrations. Error bars represent normalized standard deviations from multiple measurements.

Fig. 5. Overall collection efficiencies for polydisperse and monodisperse particles.

Field Campaign and Comparison with Parallel Filters

Good agreements between suspensions collected by PSAS and parallel filters for certain water-soluble metals were obtained. Fig. 6(a)–6(b) illustrates the comparisons for water-soluble Fe(II) and Cr(VI) between PSAS suspension samples and parallel filters. In general, agreements for both Fe(II) and Cr(VI) are observed between suspensions and parallel filters ($R^2 > 0.8$). The observed water-soluble Fe(II) and Cr(VI) concentration levels are generally in the same order compared to concentrations of water-soluble Fe and Cr of PM$_{2.5}$ reported in previous studies (Wang et al., 2015). The slopes of linear regressions for Fe(II) and Cr(VI) are 0.92 and 0.95, respectively. These slopes are consistent with the PSAS collection efficiency discussed in previous sections. In summary, agreements for certain chemical components between PSAS suspensions and parallel filters indicate that PSAS can effectively preserve chemical properties of collected particles, making it a promising technique for further deployment for online chemical and toxicological analysis of ambient PM$_{2.5}$.

It should be noted that denuders are usually recommended for PM collections involving condensational growth processes in order to get rid of soluble gaseous species (Buhr et al., 1995; Weber et al., 2001; Dong et al., 2012). In current PSAS system, denuders are not included which may result usually required. However, as discussed in previous sections, the current temperature parameters are used to restrain particle losses on tapered nozzle of the collection impactor, considering that majority of the tested aerosols are above 30 nm. For particles between 50 and 200 nm, the collection efficiencies are generally above 90%, with few particles deposited on the nozzle of collection impactor. Both polydisperse and monodisperse results indicate that in general the PSAS can effectively activate submicron particles for condensational growth, and eventually preserve the grown droplets into collected suspensions.
in positive artifacts in certain species such as volatile/semi-volatile organic compounds (VOCs/SVOCs). A previous study by Wang et al. (2013a) showed that the positive artifacts for gas-phase organics can be up to 1.5–2 µg m⁻³. Therefore, when PSAS is deployed for certain cases in which the concentration of soluble gaseous species are substantial, appropriate denuders should be added, or potential sampling artifacts of these species should be carefully evaluated. Despite these limitations, the PSAS is still adaptable for PM field sampling campaign and continuous measurements of PM chemical and toxicological analysis and monitoring.

**CONCLUSIONS**

We designed and evaluated a particle sampler for aerosol suspensions (PSAS) that directly collects aerosols as liquid suspensions. A saturation-condensation component enlarges fine particles into micron-sized droplets via condensational growth; these droplets are then sequentially collected through inertial impaction as liquid suspensions. Laboratory evaluations indicate that the PSAS possesses a collection efficiency above 85% for submicron particles, and field evaluations suggest good agreement (with linear regression slopes near 1 and R² values above 0.8) between the liquid suspensions and parallel filter samples for the analyzed chemical components. Therefore, the PSAS can be employed for online chemical and toxicological analyses of the ambient PM.

**ACKNOWLEDGEMENTS**

We acknowledge financial support from the National Natural Science Foundation of China (21707076) and the China Postdoctoral Science Foundation (2017M610928) on this study. The authors declare no competing financial interest.

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Received for review, December 4, 2019

Revised, February 6, 2020

Accepted, February 7, 2020