GENERATOR OF FINE POLYDISPERSE AEROSOL

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A simple and inexpensive laboratory generator of test aerosol is described. The generator is based on the principle of pneumatic atomization of a solution of a soluble compound by high-velocity air stream. After evaporation of solvent from produced droplets, solid particles are formed. The generator provides continuous unattended long-term operation with constant aerosol output. The performance characteristics of the generator were evaluated by spraying solutions of $NaNO_3$ and $(NH_4)_2SO_4$. The generator produced polydisperse aerosol in a fine region of particle sizes with a geometric mean diameter of 52.1 nm and a geometric standard deviation of 1.90 for the $NaNO_3$ concentration in the sprayed solution 30 g/l. The total number concentration of the produced aerosol was 3.14×10^7 cm^{-3} at a nominal air flow rate 78 l/min. The number concentration of particles increased with increasing flow rate of solution, solute concentration and pressure of compressed air through the atomizer or with decreasing total air flow rate through the generator. The change in any of these variables can be used to adjust the particle number concentration. $NaNO_3$ aerosol generation rate was 0.172 mg/min for the $NaNO_3$ concentration in the sprayed solution 30 g/l.

Keywords: Aerosol generator; Polydisperse aerosol; Fine aerosol; Nitrate; Pneumatic atomization; Nebulization.

The production of test aerosols for instrument calibration, aerosol research and the development of air-sampling equipment is an important part of aerosol technology. The aerosol generators range from relatively simple devices to complex electronic systems[1,2]. The generators like vibrating-orifice aerosol generator[3], spinning-disk aerosol generator[2], ultrasonic nebulizer[2,4] or electrospray aerosol generator[5] are often very sophisticated and rather expensive instruments, which limits their widespread use. A different approach, the atomization of solutions with compressed air in pneumatic nebulizers, provides a simple way to generate the test aerosol. However, most conventional pneumatic atomizers[1,4,6] like Collison, DeVilbiss, Lovelace, Wright or other nebulizers, suffer from unstable production of aerosol over long-time periods because of recycling of sprayed solution, which causes an increase in the concentration of sprayed solute due to sol-

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vent evaporation[2,4]. Consequently, the aerosol particle size gradually increases with time. A constant-flow liquid feed atomizer[7] avoids this disadvantage when only a fresh solution is nebulized and the liquid produced from the impacted droplets is discarded.

In this paper, a simple and inexpensive laboratory generator of test aerosol is described. It is based on the principle of pneumatic atomization of liquid by high-velocity air in a concentric nebulizer.

EXPERIMENTAL

A schematic diagram of the aerosol generator is shown in Fig. 1. The generator is composed of two main parts, the atomizer and filtered air supply. The atomizer consists of a pneumatic nebulizer and a nebulizing vessel (Fig. 2). The used concentric nebulizer is a simplified

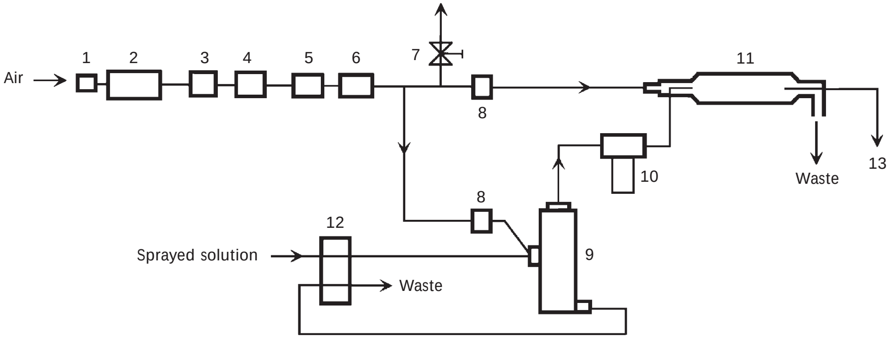

FIG. 1
Scheme of the aerosol generator: 1 coarse air filter, 2 air compressor, 3 activated charcoal, 4 silica gel, 5 fine air filter, 6 oil mist filter, 7 protection valve, 8 pressure gauge, 9 atomizer, 10 water trap, 11 drying chamber, 12 peristaltic pump, 13 standard aerosol sampling tube

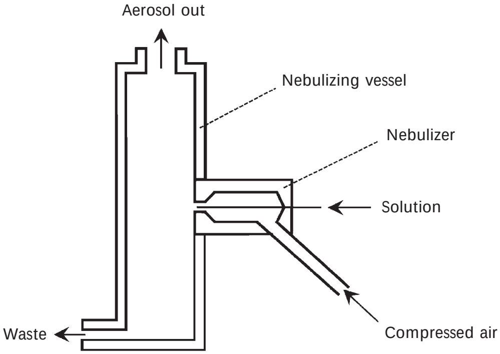

FIG. 2
Scheme of the atomizer

version of the concentric nebulizer used for the production of liquid aerosol droplets in chemiluminescent detection of nitrogen dioxide[8] and ozone[9]. The nebulizer consists of an inner stainless steel capillary (ID = 0.8 mm, OD = 1.2 mm) and an outer stainless steel coaxial tube (ID = 9 mm), which is narrowed at the end to form an orifice with a diameter of 1.4 mm. The end of the capillary is fixed centrally at the orifice. The whole nebulizer is screwed on the wall of the nebulizing vessel (stainless steel tube, ID 14 mm, length 65 mm), which is oriented vertically. A dry compressed air flows through an annulus between the inner capillary and the outer tube of the nebulizer and expands through the orifice to form a high-velocity jet. The sprayed solution delivered to the jet through capillary by means of a peristaltic pump (Ismatec, type ISM 852) is atomized at the capillary outlet by the high-speed air jet. The produced droplets are rapidly accelerated to the speed of the airstream and coarse droplets in the produced spray impact the opposite wall of the vessel and the formed liquid flows down due to gravity to the bottom of the vessel from which is continuously removed. The fine aerosol spray leaving the atomizer at the top passes through a water trap (TSI), where large droplets are collected. The spray is then mixed in a drying chamber with dry filtered air to evaporate water from the droplets to form dry aerosol particles. To analyze generated aerosol particles, the aerosol stream was sampled through a copper tube (OD 1/4″) coaxially placed at the outlet end of the drying chamber.

An air compressor (GAST, Model 71R647-P10-C322TX) is used as a source of compressed air. Air is cleaned and dried by passing through cartridges with activated charcoal and silica gel, respectively. An air filter (CKD, Model F1000-8G) and an oil mist filter (CKD, Model M1000-8G) remove particles from compressed air stream. The air stream then splits into two lines, the first one atomizing the liquid in the nebulizer while the other stream is mixed in the drying chamber (glass tube, ID 4.5 cm, length 110 cm) with the aerosol stream from the nebulizer to dry the produced droplets. The flow rate of air in both lines is regulated by means of pressure gauges (CKD, Model R1000-8G-G3). The air pressure 1 bar corresponds to the air flow rate 18 l/min through the atomizer. The flow rate of drying air is 60 l/min.

The rate of aerosol generation was measured by sampling $NaNO_3$ aerosols on a microfibre quartz filter (Whatman QM-A, 4.7 cm) at a flow rate 5 l/min. After sampling, the filter was extracted in ultrasonic bath with 8 ml of deionized water for 25 min at ambient temperature. The nitrate concentration in the filter extract was determined by means of the FIA technique with chemiluminescent detection[10]. The collection efficiency of a single filter for tested aerosol particles was 100% under the used experimental conditions.

The parameters of produced aerosol were evaluated with a scanning mobility particle sizer (SMPS; TSI Inc., U.S.A., Model 3934) by spraying solutions of $NaNO_3$ and $(NH_4)_2SO_4$. The production of particles with larger diameters was examined by means of an Aerosol spectrometer Welas 2000 (Palas, Germany) that operates in the size range from 0.3 to 17 μm.

All solutions were prepared with a distilled deionized water. $NaNO_3$ (Aldrich, Milwaukee, U.S.A.) and $(NH_4)_2SO_4$ (Lachema, Brno, Czech Republic) were of analytical grade.

THEORY

The problems of aerosol generation and relevant topics are not very frequent in this journal; therefore, the used expressions are first briefly explained.

The behaviour of aerosol particles depends strongly on their size which is defined by the particle diameter. Most aerosol particles are polydisperse, i.e. they are present in a variety of sizes. To describe simultaneously the concentration and size of individual particles present in the aerosol, a particle size distribution is mostly used[1,11,12]. The particle size distribution (by count) is formed by the fractions of the aerosol represented by number of particles in contiguous size intervals. Normal distribution, which is symmetrical, is seldom used to describe the particle size distribution because most aerosols are polydisperse, which exhibit a skewed distribution function with long tail at large sizes. The particle size distribution of polydisperse aerosol is the most frequently described with a log-normal distribution that assumes that the logarithms of particle diameter plotted on *x*-axis are normally distributed. For the log-normal distribution, the number frequency function can be written as

$$\mathrm{d}f = \frac{1}{\sqrt{2\pi}\, d_\mathrm{p} \ln \sigma_\mathrm{g}} \exp\left[-\frac{(\ln d_\mathrm{p} - \ln \mathrm{CMD})^2}{2(\ln \sigma_\mathrm{g})^2}\right] \mathrm{d}d_\mathrm{p} \qquad (1)$$

which gives the fraction of the particles having diameters whose logarithms lie between ln d_p and ln d_p + d ln d_p. Particle diameter is denoted as d_p. CMD, a count median diameter, is defined as the particle size that splits the particle size distribution (by count) into two equal halves. The log-normal distribution is characterized by the geometric mean diameter d_g and the geometric standard deviation σ_g. The geometric mean diameter, defined as the arithmetic mean of the distributions of ln d_p, identifies the centre of the log-normal distribution and the geometric standard deviation characterizes the width of the distribution. Both these parameters are usually obtained from count (number) size distribution. They are mathematically defined as

$$\ln d_\mathrm{g} = \frac{\sum n_i \ln d_i}{\sum n_i} \qquad (2)$$

and

$$\ln \sigma_\mathrm{g} = \left[\frac{\sum n_i (\ln d_\mathrm{g} - \ln d_i)^2}{\sum n_i - 1}\right]^{1/2}, \qquad (3)$$

where d_i is the midpoint diameter of a given particle size interval and n_i is the number of particles in group *i* having the midpoint size d_i and $N = \sum n_i$, the total number of particles, and summations are over all size intervals.

The geometric standard deviation serves as a criterion of aerosol dispersity; for a monodisperse aerosol, σ_g is equal to unity. For the log-normal distribution, the geometric mean diameter equals the count median diameter.

Aerosol production in pneumatic nebulizers has been studied extensively in the past[13,14]. Atomization of liquid is a complex process and although much attention has been given to study of the fundamental mechanics of liquid shattering to form small droplets, the precise mechanism of droplet formation is not completely understood yet[6,15–17]. Nevertheless, a few models have been proposed to describe the droplet formation[17]. The prevalent “surface stripping” model assumes that fine ligaments or sheets of liquid are drawn from an unstable surface of the liquid stream, as it interacts with high-velocity gas stream which passes the mouth of the liquid inlet tube. Ligaments or sheets, in turn, disrupt to give small droplets[4,17,18]. The primary droplet distribution produced by the atomization of the liquid is mostly described by the empirical equations of Nukiyama and Tanasawa[13,14,19]. The first equation gives the average droplet diameter of the produced spray in terms of operation variables of the nebulizer and the properties of the liquid being sprayed.

$$D_\mathrm{S} = \frac{585\sqrt{\sigma}}{V\sqrt{\rho}} + 597\left(\frac{\eta}{\sqrt{\sigma\rho}}\right)^{0.45}\left(\frac{1000\,Q_\mathrm{L}}{Q_\mathrm{G}}\right)^{1.5}, \tag{4}$$

where D_S is the Sauter mean droplet diameter (i.e. diameter of a single droplet with the same ratio of surface to volume as for the total sum of droplets), V is the velocity difference between air and liquid flows to the nebulizer, Q_L and Q_G are the volume flow rates of liquid and air, σ is liquid surface tension, η is liquid viscosity and ρ is liquid density. As the equation was derived empirically, it does not take into account the nebulizer geometry. The other equation describes the primary droplet distribution in liquid spray.

$$\frac{\mathrm{d}n}{\mathrm{d}x} = ax^{p}\mathrm{e}^{-bx^{q}}, \tag{5}$$

where x is diameter of an individual droplet, n is the number of droplets with diameter between zero and x in the whole sample and a, b, p and q are empirical constants dependent on the nozzle (mostly $p = 2$). Nevertheless, it is evident that liquid atomization depends not only on operation conditions like the applied pressure and air velocity, the ratio of air and liquid flow rates, surface tension, density and viscosity of the liquid but also on the size and geometry of nebulizer, viscosity and density of gas, etc.[13,18–21].

Droplets produced by primary nebulisation are rapidly fragmented by the action of the gas stream, or by collision with other surfaces within the chamber (secondary fragmentation). Moreover, the liquid deposited on chamber walls can be renebulised by the gas stream[17,18].

The pneumatic nebulizers produce a wide spectrum of droplet sizes and resulting aerosol particles after evaporation are concomitantly polydisperse. Droplet evaporation begins immediately since the vapour pressure on a curved surface is elevated[4]. If aerosol droplets contain a dissolved salt, evaporation of the solvent forms a dry salt particle in a short time interval[22,23].

RESULTS AND DISCUSSION

To characterize the performance of the presented generator, the particle size distribution and other important parameters of the produced aerosol were investigated. The particle size distribution was measured in the range 15–710 nm. Sodium nitrate and ammonium sulfate, as the representatives of the major inorganic ion species present in the PM 2.5 fraction of atmospheric aerosols (particulate matter, i.e., particles with diameters smaller than 2.5 μm), were used to test the aerosol production.

The number size distribution of produced aerosol as a function of the $NaNO_3$ concentration in the sprayed solution is shown in Fig. 3. It is evident that the concentration of the solute in the range 0.01–30 g/l affects

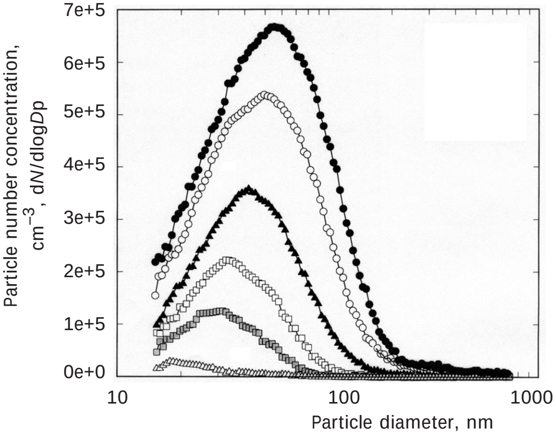

FIG. 3

Particle size distribution as a function of $NaNO_3$ concentration in the sprayed solution (in g/l): ● 30, ○ 10, ▲ 1, □ 0.3, ■ 0.1, △ 0.01. Air pressure 1 bar, solution flow rate 0.847 ml/min mainly the particle number concentration while its effect on particle size is smaller. 95% of the particles counted have a diameter under 65 and 145 nm for the $NaNO_3$ solution of concentrations 0.1 and 30 g/l, respectively. No particles are generated in the size range above 0.8 μm (measured with the Welas) even for the highest $NaNO_3$ solution concentration (i.e. 30 g/l). With increasing concentration of the nitrate in the sprayed solution, the geometric mean diameter of produced aerosol increases from 31.6 nm at 0.1 g/l to 52.1 nm at 30 g/l of $NaNO_3$. The number concentration of particles in the produced aerosol spray increases with increasing concentration of the nitrate in the sprayed solution. The total concentration of produced aerosol particles is 3.86×10^6 and 3.14×10^7 cm^{-3} for the $NaNO_3$ concentrations in the solutions 0.1 and 30 g/l, respectively, at a nominal air flow rate 78 l/min. The total particle number concentration as a function of $NaNO_3$ concentration in the sprayed solution is presented in Fig. 4. The geometric standard deviation of produced aerosol is 1.55 at 0.1 g/l and 1.90 at 30 g/l of $NaNO_3$, respectively. As it follows from these results, the generator produces the polydisperse aerosol in a fine region of particle sizes.

The effect of the liquid flow rate and compressed air pressure on the aerosol size distribution is shown in Figs 5 and 6, respectively. Both the flow rate of nebulized solution and the air pressure influence primarily the concentration of produced aerosol particles while their effect on changes in particle size is negligible. The total particle number concentration increases proportionally with the flow rate of sprayed solution while the relationship

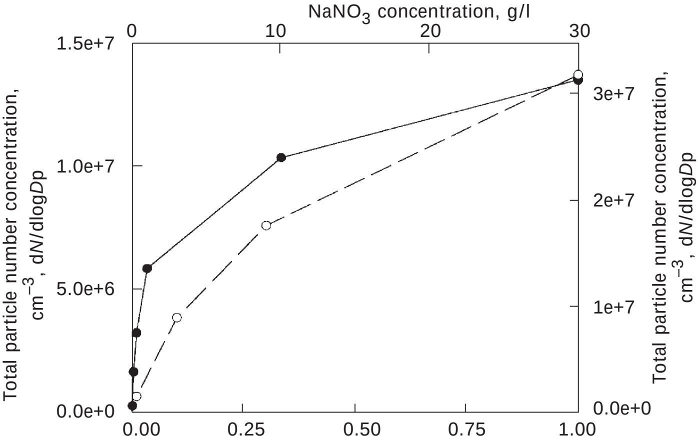

FIG. 4

The total particle number concentration as a function of $NaNO_3$ concentration in the sprayed solution: upper *x*- and right *y*-axes (●), bottom *x*- and left *y*-axes (○). Air pressure 1 bar, solution flow rate 0.847 ml/min between the total number concentration and the compressed air pressure, i.e., the air flow rate through the atomizer, is quadratic (Fig. 7).

No substantial differences in the particle size or the particle number concentration were found during the nebulization of 0.01 and 0.1 g/l of $(NH_4)_2SO_4$, as compared with the nebulization of solutions with the same concentrations of $NaNO_3$. This means that the production of aerosol in the

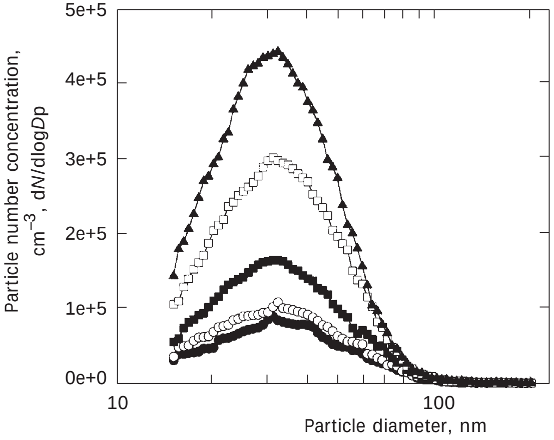

FIG. 5
Particle size distribution as a function of liquid flow rate (in µl/min): ● 211, ○ 300, ■ 443, □ 852, ▲ 1300. Air pressure 1 bar, $NaNO_3$ concentration in the sprayed solution 0.5 g/l

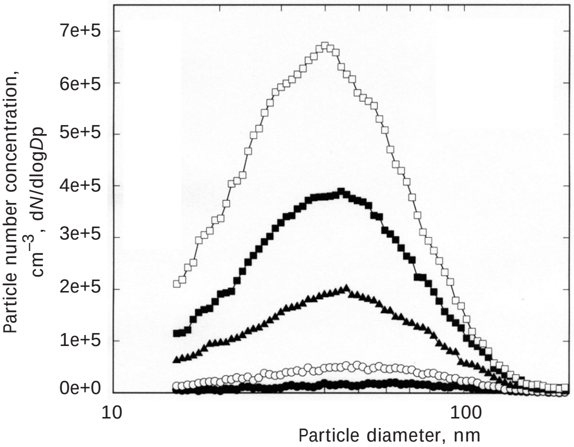

FIG. 6
Particle size distribution as a function of compressed air pressure (in bar): ● 0.25, ○ 0.50, ▲ 0.75, ■ 1.00, □ 1.25. Solution flow rate 0.847 ml/min, $NaNO_3$ concentration in the sprayed solution 1 g/l

presented generator for these two salts does not depend on the kind of salt in the nebulized solution but it is only a function of basic operation parameters of the generator, such as the liquid flow rate, air pressure and concentration of salt in the solution.

At the compressed air pressure of 1 bar and a solution flow rate of 0.719 ml/min, the $NaNO_3$ aerosol generation rate is about 0.011, 0.125 and 0.172 mg/min for the $NaNO_3$ concentration in the sprayed solution of 0.1, 20 and 30 g/l and the corresponding aerosol mass concentration at the outlet from the generator is 0.14, 1.6 and 2.2 mg/m^3 of $NaNO_3$, respectively. As it can be simply calculated, the degree of liquid atomization at the same time decreases with increasing $NaNO_3$ concentration in sprayed solution from 15.2% at the $NaNO_3$ solution concentration 0.1 g/l to 0.8% at 30 g/l of $NaNO_3$. With increasing $NaNO_3$ concentration in the sprayed solution, probably higher fractions of coarse and large droplets are produced; they are removed subsequently by impact into the inner wall of nebulizing vessel or, later on, in the water trap and, consequently, the rate of atomization decreases.

To obtain stable aerosol production, only fresh solution without recirculation of the condensed waste solution is nebulized in the atomizer. The peristaltic pump used for the solution delivery to the atomizer allows a continuous long-term generation of aerosol in comparison with a limited operation time if a syringe pump is used[7]. The Ismatec peristaltic pump combined with a flow RC regulation (resistor-capacity couple), which acts

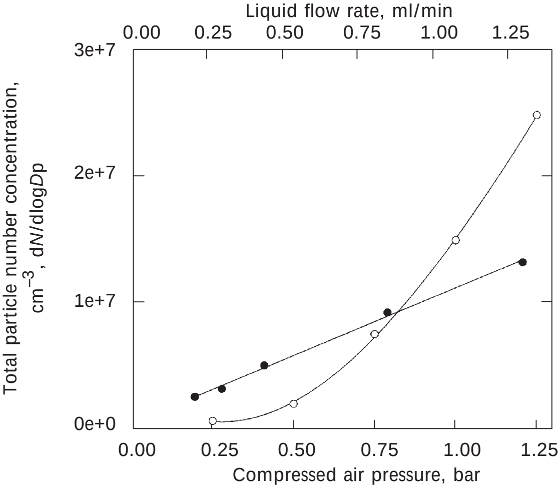

FIG. 7

The total particle number concentration as a function of compressed air pressure (○) and liquid flow rate (●)

as an effective pulse damper, provides stable, pulse-free flow of nebulized liquid to the atomizer. The stability of aerosol production in the presented generator was tested by spraying a solution of 10 g/l of $NaNO_3$ at a flow rate of 0.719 ml/min and the compressed air pressure of 1 bar continuously for 5 h. The generator output was stable over the whole measurement period although a small fluctuation in aerosol concentration (± 3% from the average value) was observed.

A low inner volume of the atomizer ensures a short response time (3–4 min) of the generator to obtain a steady-state aerosol production after a stepwise change in a basic system parameter, e.g. the solute concentration in the sprayed solution, solution flow rate and/or the compressed air pressure. A prefabricated system and a portable source of pressurized air presented in this paper allow fast and simple transportation and assembling of individual components of the generator and thus easy portability of the whole system. Combination of simplicity, flexibility and easy operation offers using the presented generator as a useful inexpensive device for the laboratory generation of standard aerosol. The total price of all used components is more than one order lower than that for commercial generators with similar parameters[24,25].

CONCLUSIONS

A stable generator of test aerosol based on the principle of pneumatic atomization of liquid by high-velocity air is described. The generator produces reproducibly a fine polydisperse aerosol with high, constant and adjustable particle concentration. Particle concentration can be changed by altering the flow rate of nebulized solution or the solute concentration in the nebulized solution or, alternatively, by changing the pressure of air flowing through the atomizer or changing the total air flow rate through the generator. Simple, flexible and easy operation suggests its use as a useful inexpensive device for laboratory generation of standard aerosol.

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