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Diffusion aerosol spectrometer

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Abstract

The diffusion aerosol spectrometer for the measurements of particle size spectra and concentration levels is described. It includes three principal parts: (i) a block of diffusion batteries for measuring the particles, whose size does not exceed 0.15 μm , (ii) the particle amplifier for growing the particles passing through the diffusion batteries up to optically distinguishable sizes and (iii) the laser aerosol spectrometer, which counts the amplified particles and may also serve for independent measurements of particle size spectra within submicron size range. The tandem including: diffusion batteries + laser aerosol spectrometer allows for detecting particles of radius > 3 nm at maximal concentration up to 2×10^4 particles/cm³. The tandem is managed either by PC or manually. The instrument is designed for studying aerosols in the atmosphere and for ecological measurements. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Although more than 40 years (and maybe more) have passed since the importance of highly dispersed aerosols (whose size is less than 10^3 nm) was well understood (Fuchs, 1964), not many instruments appeared for measuring their microphysical characteristics, particularly their size distribution. Diffusion and electromobility spectrometers start the scarce list of these instruments. Despite the latter yielding a more detailed information on the particle size spectra, the former is still widespread because of its simplicity and availability (it is not so expensive and, in principle, may easily be reproduced even in not so very powerful laboratories). Such a spectrometer constructed and built in the Laboratory for Physics of Aerodisperse Systems of the Karpov Institute participated in the intercalibration workshop that was held at the Institute for Experimental Physics of the University of Vienna.

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No aerosol instrument measures the particle size directly (unless the particles are sufficiently large). Instead, the measurement procedure include two steps: (1) measuring the instrument response from a single particle (or a population of particles), and (2) restoring the particle size spectra from the instrument readings.

In the case of diffusion aerosol spectrometer, the instrument response is the ratio of outlet and inlet particle concentrations dependent on the particle size. Even this ratio is still unavailable from direct measurements, because the aerosol particles are too small to be directly detected. The amplification block allowing the particle enlargement up to optically visible size is therefore an integral part of any diffusion aerosol spectrometer. The enlarged particles are then detected by a counter (the laser aerosol spectrometer in our case).

The data from the laser counter should then be processed and inverted into the particle size distribution. This step is the most critical one, for the data inversion in this case belongs to the class of ill-posed problems and can produce huge errors in the final results (see e.g. Farzanah et al., 1985). Therefore, we avoid the full inversion procedure and restore only the mean particle radius, the width of the size distribution and the number concentration. This can be done more or less reliably.

2. Principles of operation

Fig. 1 shows the principle arrangement of our diffusion battery spectrometer. The carrier gas containing aerosol particles enters into the diffusion battery block (6) and passes through one of five screen batteries installed in a rotating cartridge. This allows us to perform the measurements on each battery beginning with the “zeroth” one imitating the losses in the inlet and outlet parts of the batteries. A part of the entering aerosol particles deposits on the grid screens inside the batteries. The rest reaches the amplifying blocks (7 and 8) consisting of two condensational amplifiers enlarging the particles by two-step condensation up to submicron size (0.2–0.8 μm). These particles are then detected by the laser aerosol counter (11). The personal computer (10) controls the flow rate, the time of measurement exposing each battery for a given time. Furthermore, it processes the data by inverting the particle penetration ratio as a function of the number of screens into characteristics of the particle size distribution. The size distribution is retrieved from the integral equation:

$$P(n) = \int_0^{\infty} p(n, D) \cdot f(D) \cdot dD \quad (1)$$

where $P(n)$ is the measured penetration ratio, D is the aerosol particle diffusion coefficient, $p(n, D)$ is the penetration ratio for the monodispersed aerosol passing through the diffusion battery containing n screens, and $f(D)$ is the distribution of the aerosol particles over their diffusion coefficients. Once Eq. (1) is solved and the function $f(D)$ is found, the particle size distribution is readily restored:

$$F(a) = f(D(a)) \cdot \frac{dD}{da} \quad (2)$$

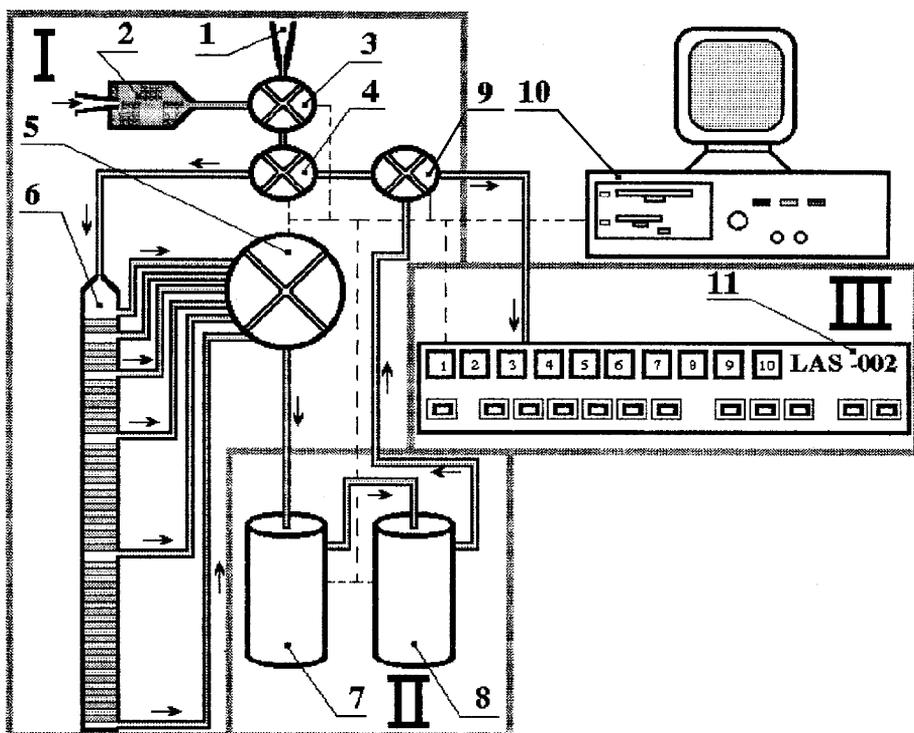


Fig. 1. Schematic diagram of the diffusion aerosol spectrometer. (1) Aerosol inlet, (2) filter (3, 4, 5) valves, (6) diffusion batteries, (7) particle amplifier with dioctylsebacate, (8) particle amplifier with dibutylphthalate, (9) valve, (10) computer, (11) laser aerosol spectrometer. I—Block of diffusion batteries; II—Particle amplifier; III—Laser aerosol counter.

where a is the particle radius and the dependence of the diffusion coefficient on a is assumed to be known.

The solution of Eq. (1) turns out to be difficult, for even small experimental errors in $P(n)$ result in huge errors in the particle size distribution. We therefore restrict ourselves by determining only the position and the width of the size distribution. These two parameters are found from the solution of nonlinear equations arising when the integral in Eq. (1) is evaluated by the saddle point method (details can be found in Lushnikov and Zagaynov, 1990).

Fig. 2 shows the dependencies of $\ln P(n)$ on the number of screens in the battery for several lognormal size distributions. Typically, these curves approach straight lines at $[\langle D \rangle / (\sigma)] \rightarrow \infty$, where $\langle D \rangle$ is the averaged particle diffusion coefficient and σ is the width of the distribution $f(D)$. We performed several numerical experiments and found that $P(n)$ may be approximated by the function:

$$P(n) = \exp\left(-a_1 \langle D \rangle^{\frac{2}{3}} \cdot n + b \cdot \sigma \cdot n^{\frac{3}{2}}\right). \quad (3)$$

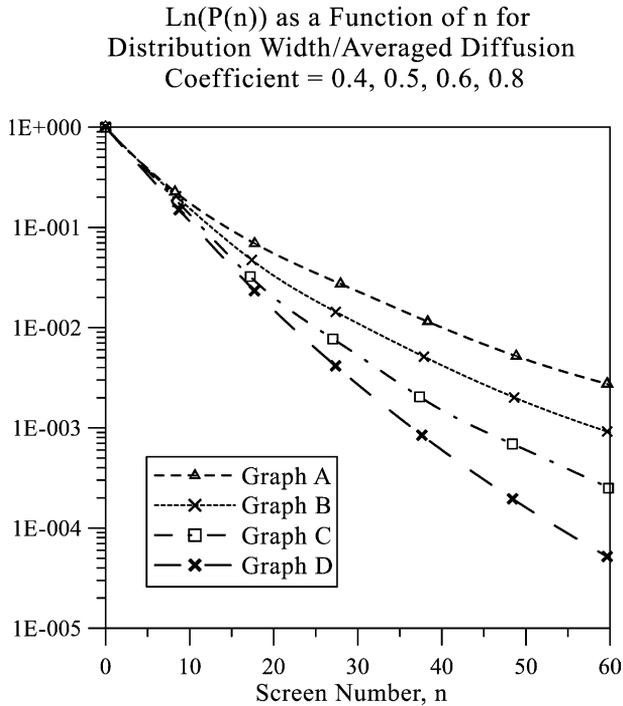


Fig. 2. Penetration coefficient as a function of the screen number for different distribution widths: $\sigma/\langle D \rangle = 0.4; 0.5; 0.6; 0.8$.

The coefficient $a_1 = A(2r_0u_0)^{-2/3}$, $b = 23.7$; here r_0 is the fiber radius, u_0 is the undistributed flow velocity and A is a constant (see Cheng and Yeh, 1980).

Eqs. (2) and (3) serve for the computer processing of the experimental data. Each measurement thus gives three parameters: particle number concentration, the position of the maximum size distribution and its width.

3. Technical description

3.1. Block of diffusion batteries

Consists of five diffusion batteries containing 0, 2, 5, 10 and 20 screens. This choice provides the most effective determination of the parameters of the size distributions of highly dispersed aerosols whose particle size varies within 1–100 nm, the ratio $\langle D \rangle / (\sigma)$ varies from 0.1 to 0.9, and the number concentration n ranges from 10 to 10^6 cm^{-3} . The batteries are installed in a rotating cartridge allowing the aerosol to pass through each battery in turn. The exposition time depends on the aerosol concentration and is chosen by the PC (10) in Fig. 1. Typically, it does not exceed 1 min. The volume flow rate also chosen by the PC varies from 2 to 4 l/min.

3.2. Particle amplifier

Consists of two stages, the first of which enlarges the particles up to 30 nm by condensing in the saturated dioctylsebacate vapour onto their surfaces. The second stage filled with saturated dibutyl phthalate vapour allows the particles to reach their final size 0.3–0.4 μm . The two-stage amplification has some advantages: it allows for more flexible control of the outlet particle size and increases the thermal stability of enlarged particles, providing their further transport with no loss. It is very important to be sure that: (i) all particles are enlarged, and (ii) the size of all enlarged particles exceeds the sensitivity threshold of the laser aerosol counter. To this end, the approach proposed by Kousaka et al. (1985) is used:

$$\Delta H = \frac{\pi}{6} \cdot (d_f^3 - d_i^3) \cdot n \cdot \rho_s \cdot v_H \quad (4)$$

where d_f =final particle diameter, d_i =initial particle diameter, n =particle number concentration, v_H =specific volume of humid air, ρ_s =density of condensed liquid, and ΔH =quantity of condensable vapour in a unit mass of dry air.

Since $d_f \gg d_i$, Eq. (4) may be cast into the form:

$$d_f = \left(\frac{6 \cdot \Delta H}{\pi \cdot n \cdot \rho_s \cdot v_H} \right)^{\frac{1}{3}}. \quad (5)$$

It means that d_f does not depend on d_i . Moreover, Eq. (5) only illustrates the situation, because the main part of ΔH deposits on the walls of the amplifier and a much smaller part of vapour condenses onto the particle surfaces. This means that the final particle diameter depends on neither initial diameter nor particle concentration.

Generally speaking, this amplifier can work in the regime when its sensitivity is as low as 1 nm (see Zagaynov et al., 1976). In the present case, the amplifier activates the particles larger than ≈ 3 nm.

3.3. Laser counter

The in-cavity laser aerosol spectrometer (see Kirsch and Julanov, 1978) is used in our setup as the particle counter. The counter sensitivity is 0.15 μm by particle diameter at maximal particle number concentrations up to 2×10^4 particles/cm³. The spectrometer can be used independently for the aerosol measurements within the submicron size range.

The detection probability of the optical counter $\eta(x)$ may be evaluated from the following consideration. As was shown in Kirsch and Julanov (1978), the laser beam is wider than the airflow carrying aerosol particles crossing the laser beam. This means that all the particles in the flow are counted once the scattered signal exceeds the sensitivity threshold of the spectrometer. Therefore, $\eta(x)$ may be expressed in terms of the Heaviside step function $\theta(x)$:

$$\eta(x) = \theta(r_0) \quad (6)$$

with r_0 being the laser aerosol spectrometer sensitivity ($2r_0 = 0.15 \mu\text{m}$ in our case).

3.4. Management and control

Special software was developed for treating the experimental data, creating a data bank and controlling the diffusion aerosol spectrometer. The PC controls the temperatures of the amplifiers, the airflow rate, rotates the cartridge with the diffusion batteries and chooses the time for a single measurement. Special regimes with a changed configuration of the spectrometer may be used. The maximum time for a measurement is 5 min.

4. Calibration

4.1. Calibration of the diffusion batteries

To this end, a monodispersed aerosol of known concentration and particle size should be passed through each battery and the penetration ratio measured for a set of values of these parameters. Because the construction of our batteries does not differ from that of Cheng and Yeh (1980), we used the calibration data from this article.

4.2. Calibration of the size amplifier

We measured particle losses in both stages and found them to be less than 20%. The size of the particles enlarged by the first stage of the amplifier was estimated theoretically (it is not of great importance for our final goals). The second stage was calibrated with the aid of the laser aerosol spectrometer. The measurements showed that the two-step enlargement produces rather monodispersed aerosols, the parameters of which are: $\langle r \rangle = 0.4 \mu\text{m}$, $\langle r \rangle / (\sigma) = 0.2$.

These characteristics depend on the type of measured aerosol and change within a range of 0.2–0.9 μm . No vapour depletion effects were observed up to the number concentration of 10^6 particles/cm³.

4.3. Calibration of the laser aerosol spectrometer

We did not deviate far from the standard procedure of the calibration of optical aerosol spectrometers described elsewhere (see, e.g. Kirsch and Julanov, 1978) and calibrated our spectrometer by a set of latex aerosols provided by Dow Chemical. It was found that the instrument measures the particle size within the size interval (0.15–2 μm) at a maximum concentration of 2×10^4 particles/cm³. The latter is limited by coincidence losses.

5. Measuring range and limitations

The diffusion battery aerosol spectrometer measures the position of the maximum and the width of the particle size distribution of aerosols whose particle size varies within the size range of 3 nm–1 μm . Listed below are the limitations of the instrument.

(i) The instrument measures only two first moments of the size distribution. Attempts to get more information fail due to the necessity to solve the ill-posed inversion problem.

(ii) The concentration limitation (upper limit) is caused by the effects of multiple counting in the laser aerosol spectrometer. This limitation is easily relaxed either by using a diluter or by application of a modified theory taking these effects into account (Julanov et al., 1984).

(iii) The origin of the size limitations needs no explanations.

(iv) The time needed for a single measurement is about 5 min or 1 min per one battery. This fact causes an additional limitation related to the time fluctuations of the particle size distribution, which are able to distort completely the information on the size spectrum.

6. Main applications

The diffusion aerosol spectrometer is designed specially for measurement of the characteristics of atmospheric aerosols. We were able to use it for the measurements of background aerosols (Lake Baikal, Zagaynov et al., 1989), in the forests of East Lithuania, Caucasus mountains (Zagaynov et al., 1987) and in rather contaminated industrial regions (Gomel, Bratsk). However, the instrument can be also used for some technological and scientific measurements.

7. Summary and conclusion

During the intercomparison workshop in Vienna, we had a good opportunity to compare the results of our measurements with those given by other instruments. As previously, it was found that our instrument produces reasonable aerosol characteristics, showing that the diffusion battery method is applicable for measurements of parameters of highly dispersed atmospheric aerosols.

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