Active Spectral Nephelometry in Studies of the Condensational Activity of Submicron Aerosol

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Introduction

Water vapor condensation and evaporation are among the main processes of the atmospheric aerosol transformation essentially affecting its optical and radiative characteristics. Most of the known methods for investigating the aerosol condensation activity are based on measurements of only the changes in the particle size with relative humidity (RH). Variation of the particle refractive index in these cases can be estimated only by means of model calculations.

We have developed another approach to investigations of the aerosol properties and processes of their transformation based on measurements of the directed scattering coefficient in visible wavelength range by means of a FAN nephelometer using artificial humidification (from 20% to 90%) and heating (up to 300°C). So the method can be called “active spectral nephelometry.” All measurements are fulfilled for natural aerosol sampled from the atmosphere.

As applied to the study of aerosol condensation activity, the method implies measurement of optical characteristics of ambient aerosol sampled from the atmosphere, during which the particles are subjected to artificial humidification.

Approach

The change of the scattering coefficient and also directed scattering coefficient (or its polarized component) as RH increases can be described by the empirical Kasten–Hanel formula:

$$\sigma = \sigma_0(1-\mathrm{RH})^{-\gamma}, \tag{1}$$

where σ is the scattering coefficient, σ_0 is the scattering coefficient due to the dry matter of aerosol particles, and γ is the parameter of condensation activity.

Measurements of the only directed scattering coefficient at the angle of 45° and at the wavelength of 0.5 µm were carried out early in our investigation. Later the method was developed when passing to measurements of the directed scattering coefficients at three wavelengths (0.41, 0.5, and 0.63 µm) at the angle of 45° and two its orthogonal polarized components at two wavelengths (0.45 and 0.51 µm) at the angle of 90°. The example of the directed scattering coefficient at 45° and two its orthogonal polarized components at 90° measured on August 12, 2001, are shown in Figure1 as functions of ln(1–RH). All other illustrations concerning the effect of RH on the aerosol characteristics are related to this date.

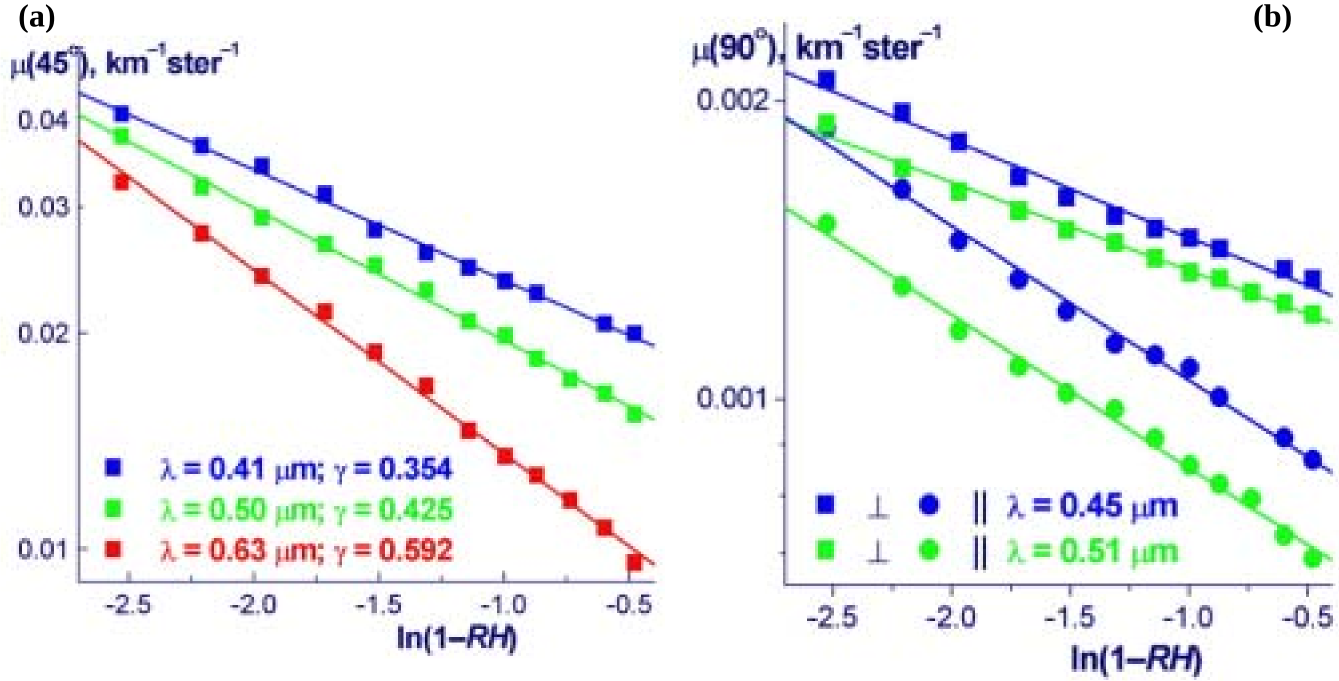

Figure 1. Directed scattering coefficient at 45° (a) and two orthogonal polarized component of scattered radiation at 90° (b) as functions of ln(1–RH) and their approximation by the dependence of the type of the Kasten–Hanel formula.

Then the data were inverted to the size distributions in the size range 0.07 to 0.9 µm. In this case solution of the inverse problem makes it possible to determine simultaneously the dynamics of the size spectrum and the complex refractive index. At the same time, the limited wavelength range and small set of measured characteristics, based on which the inverse problem was solved, requires careful study and correct determination of both the boundaries of the retrieved size spectrum and the errors in estimation of the refractive index.

Results

Figure 2 shows how the volume size distribution function changes as RH increases. The change in the total volume of aerosol particles and their refractive index as RH increases is shown in Figure 3.

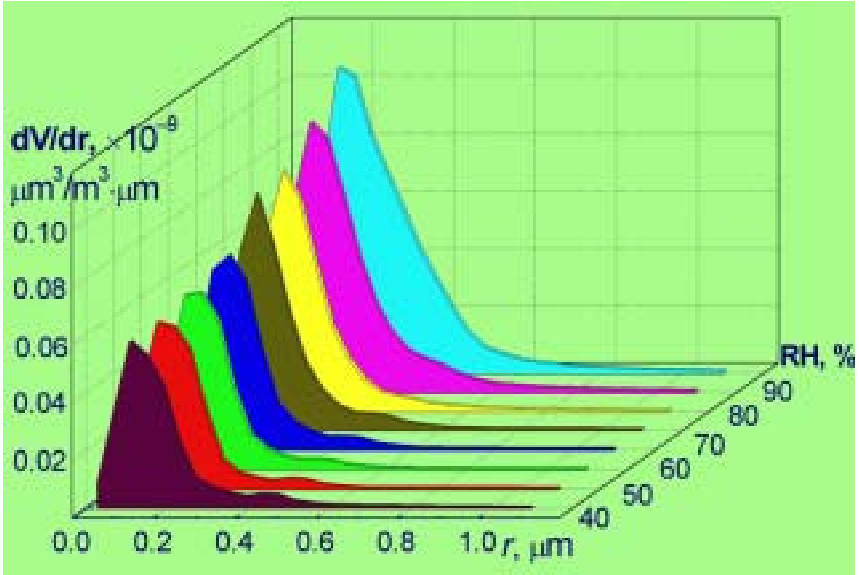

Figure 2. The volume distribution function as function of RH.

The growth factor of aerosol particles in the range of RH from 40% to 90% was also determined:

$$GF = \frac{r(RH = 90\%)}{r(RH = 40\%)} \tag{2}$$

where r is the radius of aerosol particle.

A similar approach to the analysis of thermograms provides for the data on the concentration of the volatile substances and their size distribution. The idea to apply the technique of thermoanalysis, widely used in analytic chemistry to investigate properties of aerosol particles, was developed by Lubovtseva and Yudin (1979) for optical measurements under atmospheric conditions (thermooptical method). At the same time, measurements of the scattering coefficient or the directed scattering coefficient at one wavelength (Panchenko et al. 1995) allow only to qualitatively estimate the transformation of the total volume of submicron particles with heating. Measurements of the seven parameters of the scattered radiation (the same as were measured when studying the condensation transformation of aerosol microstructure) make it possible to assess the transformation of the aerosol size spectrum and the refractive index at artificial heating of aerosol particles up to 250°C. The “volatility factor” was introduced analogously to the growth factor of aerosol particles (see Figures 3-6):

$$\mathrm{VF} = \frac{r(T = 20°C)}{r(T = 100°C)} \tag{3}$$

To test the algorithm for solving the inverse problem and to avoid uncertainties in estimates of the growth factor caused by a priori unknown range of the size spectrum, which can be retrieved from optical data, the controlled cut-off of aerosol is used. The 4–stage diffusion battery of a screen type was applied for the cut-off on the lower limit of particle size.

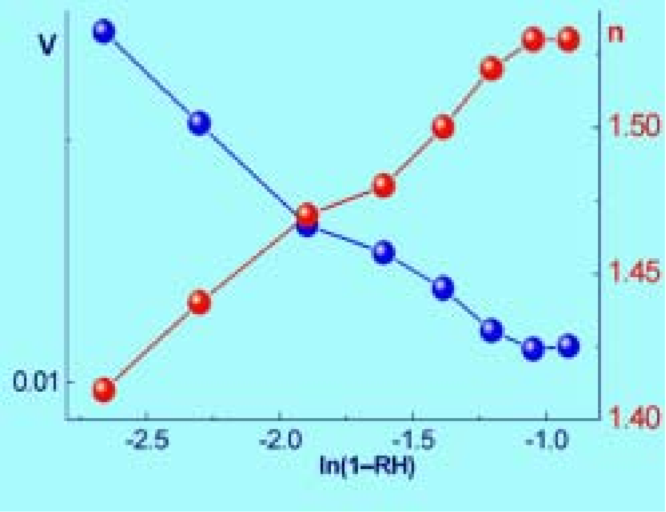

Figure 3. The total volume of aerosol particles and their refractive index as functions of RH.

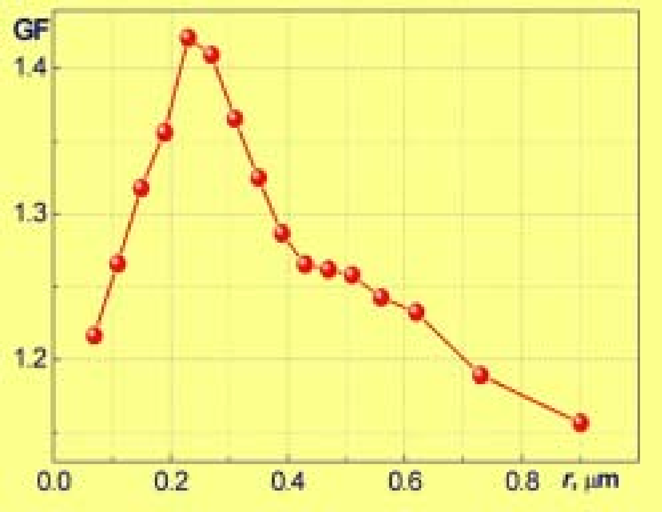

Figure 4. Growth factor of aerosol particles in the range of RH 40 to 90%.

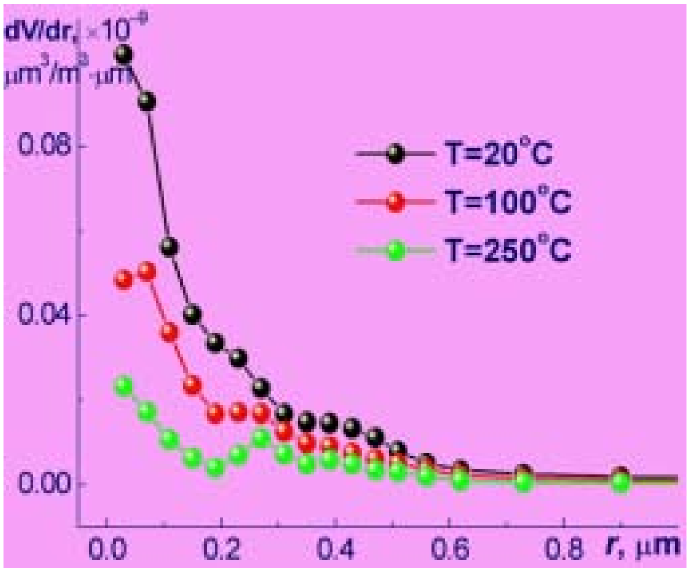

Figure 5. The change in the volume distribution function at heating from 20° to 250°C.

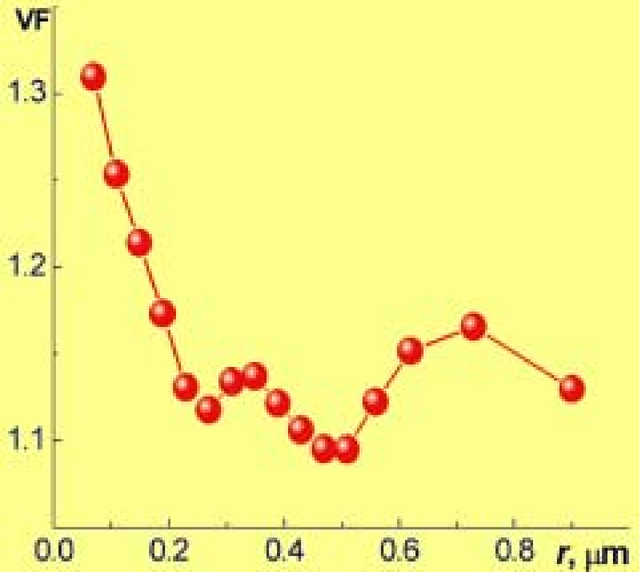

Figure 6. Volatility factor of aerosol particles in temperature range 20° to 100°C.

The change in the particle volume distribution at cut-off by different screens of the diffusion battery is shown in Figure 7a. Figure 7b shows the penetration coefficient of different screens of the diffusion battery (calculation and measurement).

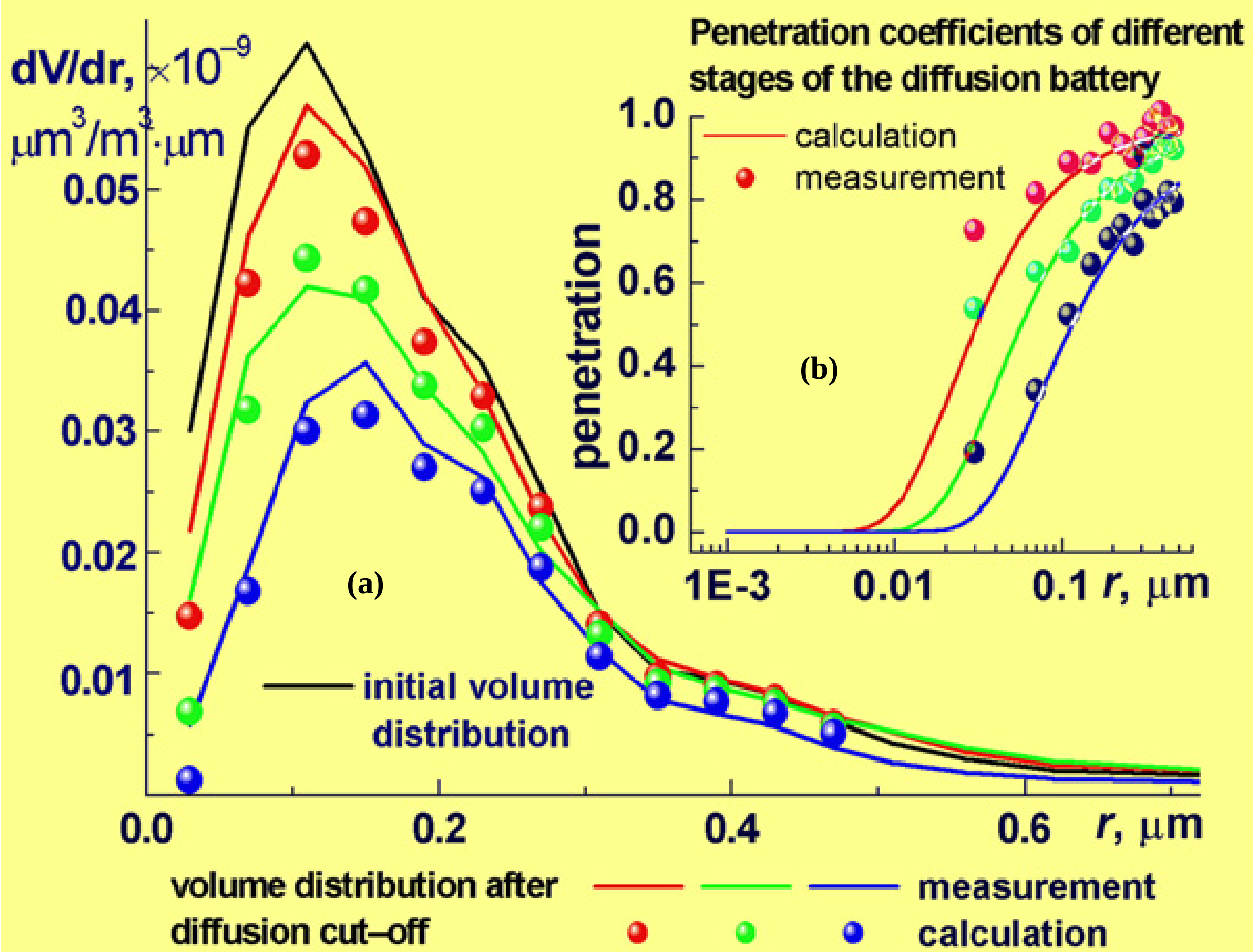

Figure 7. The change in the volume distribution function at cut-off using different stages of the (a) diffuse battery and (b) penetration coefficients of different screens of the diffusion battery.

Conclusion

The developed technique for measurement of seven parameters of the scattered radiation at heating of ambient aerosol and subsequent solution of the inverse problem opens up fresh opportunities for the study of thermal transformation of aerosol by optical methods, in particular, of the change in the particle size distribution and the refractive index at different stages of artificial heating of real aerosol.

The results obtained are evidence of the high efficiency of the method and, in our opinion, can be important for correct setting the atmospheric aerosol optical characteristics when interpreting the results of comprehensive radiative experiments.

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