Spectronephelometric Study of the Aerosol Transformation with Artificial Change of Relative Humidity

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Introduction

Water vapor condensation and evaporation is one of the main processes of the atmospheric aerosol transformation, significantly affecting its radiative characteristics. Models of the aerosol optical properties are usually based on the assumption of the similar change of the radius of the particles of certain aerosol fraction with relative humidity. Size dependence of the aerosol condensation activity earlier was studied using impactors and optical microscope (Meszaros 1971) and photoelectric aerosol counters (Laktionov 1988). Both methods are unsuitable for particles with radii $r \leq 0.2$ mcm. In the present study, we made an attempt to investigate the possibility of the evaluation of the dependence of the particle response on the variation of the relative humidity upon dry particle size with help of the nephelometric and polarimetric measurements.

Instruments

Measurements were carried out synchronously by two spectronephelometers elaborated in the Institute of Atmospheric Physics, Russian Academy of Sciences. The first nephelometer measured directed scattering coefficients for a scattering angle of 45° in the spectral range 254 nm to 578 nm with a mercury discharge lamp as the light source. Emission lines were separated by a spectrometer with diffraction lattice. A halogen filament lamp was used in the second nephelometer. Measurements were made in four spectral ranges from 0.44 mcm to 0.8 mcm separated by filters of colored glass. The measurement parameters were directed scattering coefficients at scattering angles of 45° and 90° and degree of linear polarization at scattering angle of 90°. The air passing through the chambers of nephelometers could be heated to the temperature exceeding the ambient for approximately 15° C to obtain the optical characteristics of the dry aerosol particles. Temperature and relative humidity of the outdoor air and temperature of the air flow in nephelometers were controlled.

Method

Assuming an unambiguous relation between sizes of dry and moist particles, the ratio of their radii r_w/r_d (i.e., the growth factor) can be found as the ratio of radii corresponding to the same value of the integral particle size distribution N(r) (Laktionov 1988). Integral size distribution is determined as

$$N(r) = \int_{r}^{r_{max}} n(r)dr \qquad (1)$$

where n(r) is differential size distributions.

Retrieval of the aerosol size spectra from nephelometric and polarimetric measurements is a complex problem, because kernel functions, depending upon refractive index, are unknown a priori and must be determined using measurement data. Our inversion technique was based on the iterative algorithm of Twitty (1975). Additional weighted logarithmic smoothing was included into the iteration procedure. A set of kernel functions was calculated for the real part of the index of refraction m within m = 1.32 - 1.6 and imaginary part k within k = 0 - 0.015. The refractive index and corresponding size distribution were chosen using the criterion of the minimal averaged relative r.m.s deviation between measured and recalculated optical characteristics. The applicability of the inversion technique was tested on model size distributions. The example of the model retrieval of the refractive index are presented in Figure 1 for exact input data (1) and disturbed by 5% random error close to the experimental errors (2).

Figure 2 shows the results of the retrieval of the model lognormal size distribution with median radius r_m = 0.05 mcm, variance of the logarithm of radius ν^2 = 0.6, and m = 1.45, the same as in Figure 1. Curve 1 in Figure 2 is initial distribution, Curve 2 is retrieved from exact input data, Curve 3 corresponds to the 5% error, and Curve 4 corresponds to the erroneous input data false m = 1.45. Model simulations showed that refractive index can be estimated from our experimental data with accuracy $\Delta m \approx 0.02$ to 0.03 and $\Delta k \approx 0.005$ and size distributions can be retrieved in the radius range 0.07 mcm to 1.1 mcm.

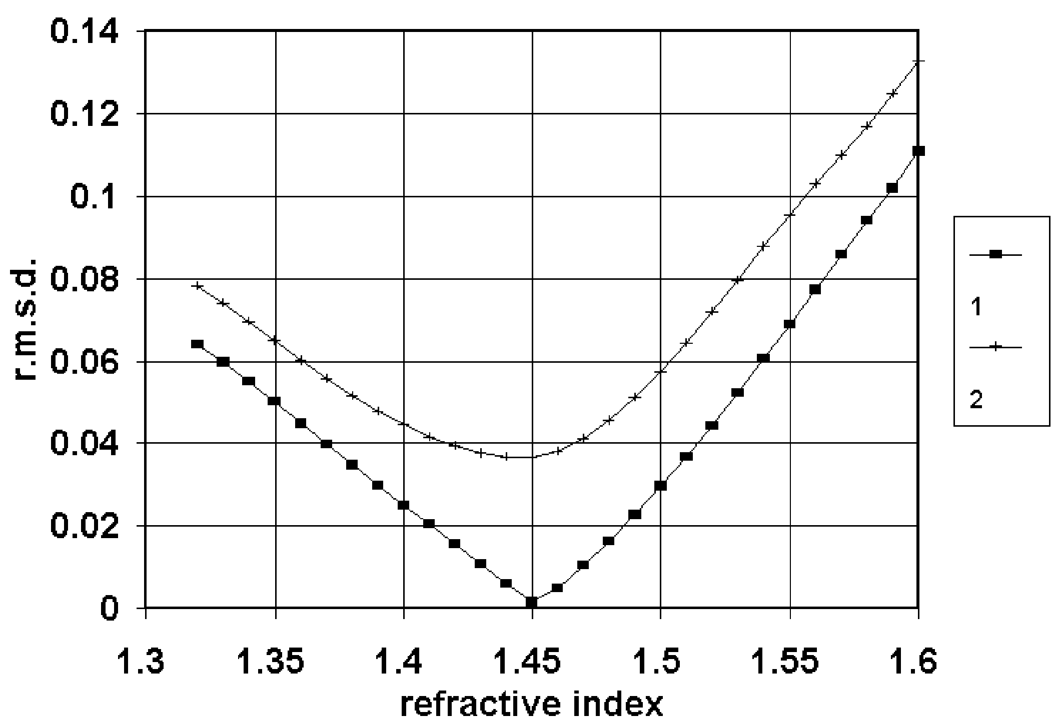

Figure 1. Estimation of the refractive index.

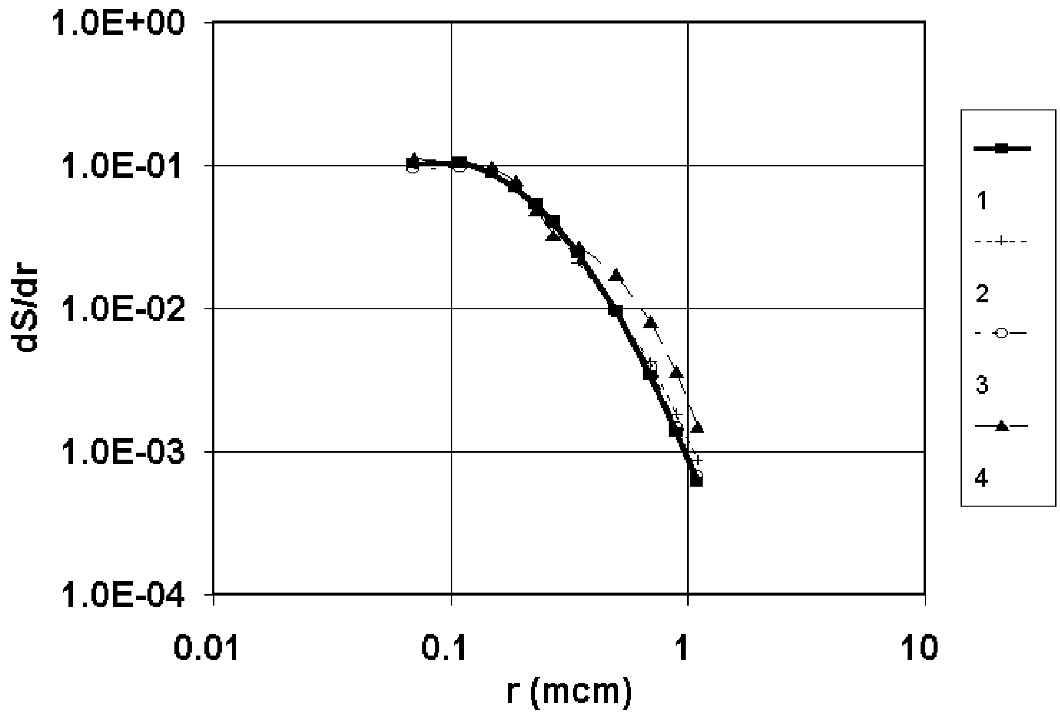

Figure 2. Retrieval of the model size distribution.

For the estimation of ratio r_w/r_d, the spline interpolation of the r_w as a function of the $N_w(r)$ in the points corresponding to $N_d(r_i)$, where r_i - radii for which N(r) were retrieved, was used.

Results

For the estimation of the aerosol condensation transformation, data were analyzed from our measurements at the Zvenigorod, Moscow region in January and February 1986 and in Odessa at the Black Sea shore in September and October 1987. Situations with a big initial relative humidity of RH > 70% were selected. After heating, it decreased to RH ≈ 20% to 30%. The estimated imaginary part of the refractive index did not exceed k = 0.005. The examples of the retrieved $N_d(r)$ and $N_w(r)$ are given in Figures 3 and 4.

Figure 5 presents the dependencies of r_w/r_d upon r_d. It can be seen from Figure 5 that the growth factor has a maximum in the size region 0.2 mcm to 0.3 mcm in most cases.

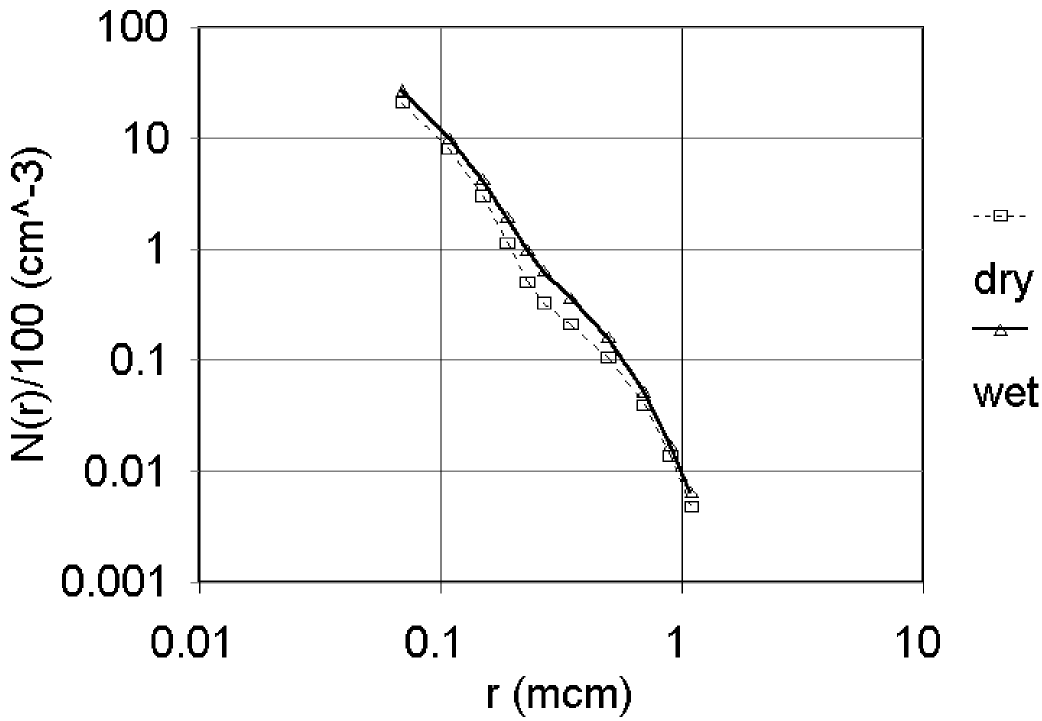

Figure 3. Integral size distributions of dry and natural aerosol. Avenigorod, 12.02.86.

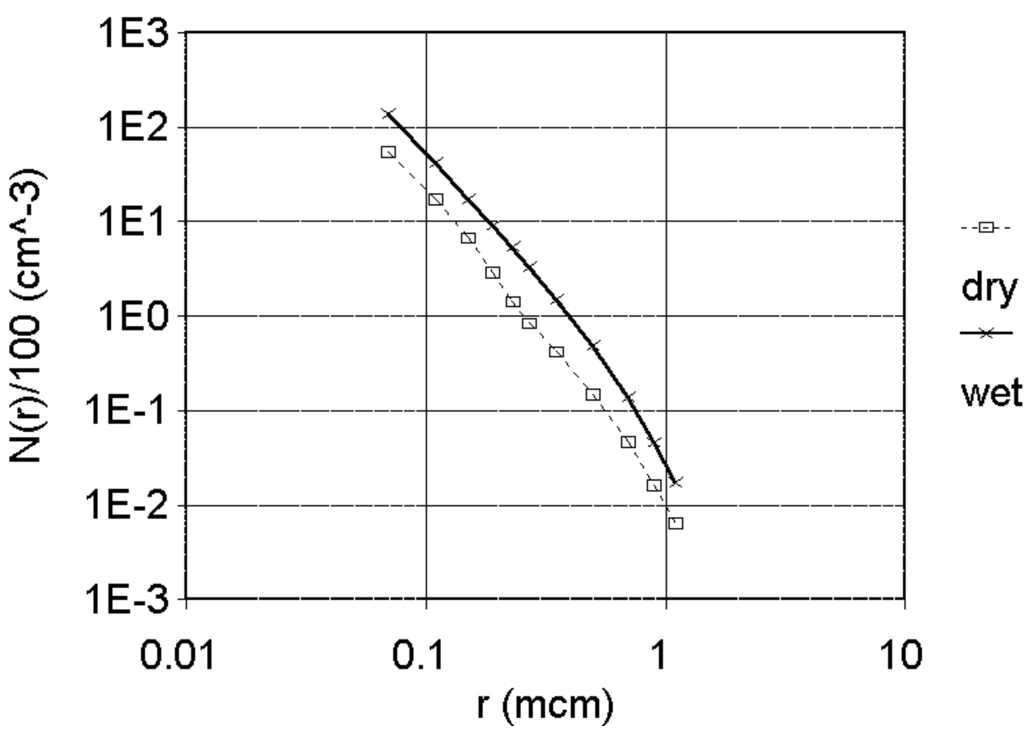

Figure 4. The same as in Figure 3. Odessa, 27.09.87.

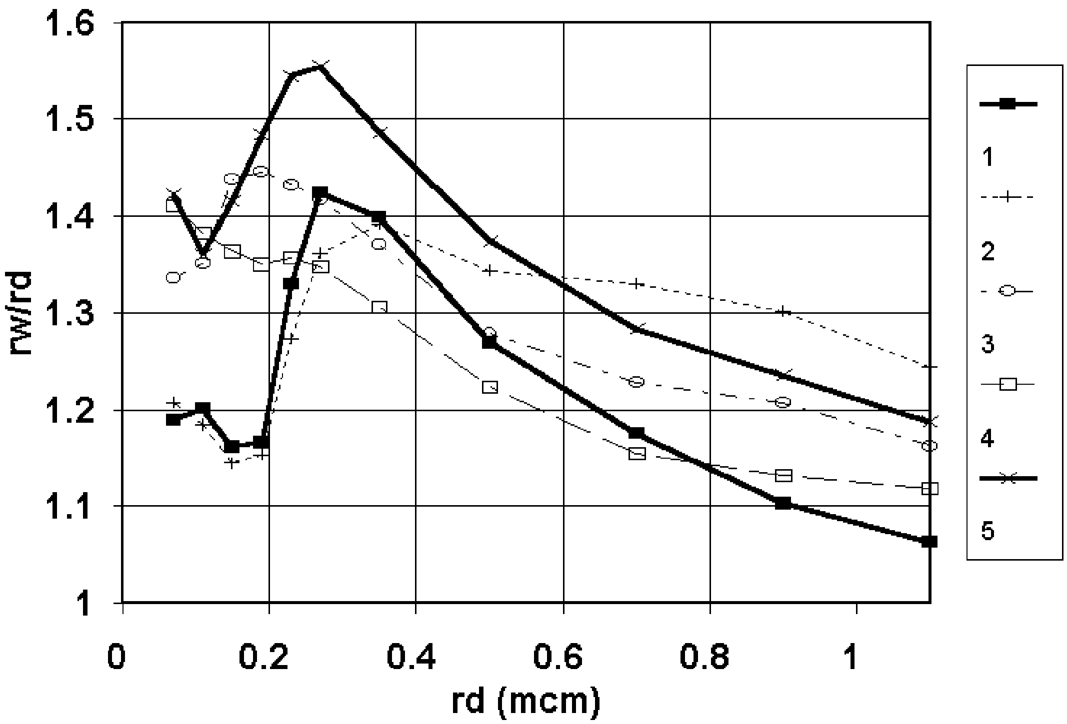

Figure 5. Dependence of the growth factor upon dry particle size. 1, 2 – Zvenigorod, January 1986; 3, 4, 5 – Odessa, September and October 1987.

Discussion

For particle radii greater than 0.4 mcm, our data do not contradict the results of Meszaros (1971) and Laktionov (1988). The values published by Laktionov of r_w/r_d for relative humidity RH = 90% are equal to 1.32, 1.27, and 1.23 for dry particle radii 0.4, 0.5, and 0.63 correspondingly and close to our results. The displayed decrease of the growth factor for particles less than 0.2 mcm is not inconsistent with data of the dependence of the volume part of the soluble material in aerosol particles. It must be mentioned that the term due to the surface tension and particle curvature in the equation of the particle condensation equilibrium is insignificant for particles greater than 0.1 mcm and cannot be responsible for this effect.

The present study must be regarded as only the first attempt in the investigation of the particle response on the change of air humidity, since several problems were not considered. One of these problems is the possible size dependence of the refractive index due to the different fraction of soluble matter in particles of different sizes. However, the constant fraction would lead to the constant growth factor. Another problem is s connected with the use of the model of the homogeneous spherical particles in Mie calculations. These problems will be the subject of future study.

References

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