Study of Characteristics of Near-Surface Aerosol by Means of Spectropolarimeter and Nephelometer in the Winter Complex Experiment

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Some preliminary results are presented of surface layer aerosol investigations obtained during the 1999 winter complex experiment by means of a nephelometer and spectropolarimeter. The nephelometer measured round the clock values of directed scattering coefficient D at scattering angle $\varphi = 45°$ and wavelength $\lambda = 0.54$ μm. This value is well known to be connected with mass concentration of submicron aerosol M. The nephelometer data gave the time scanning of the total aerosol content irrespective to its qualitative characteristics. The second instrument measured the spectral dependencies of the two polarized components of directed scattering coefficient. The light source is based on a grating monochromator with a spectral region of 0.4 m to 0.75 m and a spectral resolution of about 10 nm. Three photometers with photomultipliers as photodetectors are mounted on the aerosol chamber at scattering angles $\varphi = 45°$, 90°, and 135°.

The instrument is operated by computer and its software allows performing both quasi-continuous and discrete spectral scanning. Two polarized components of directed scattering coefficient with polarization vector parallel $D_{||}$ and perpendicular $D_{\perp}$ to the scattering plane are measured. The so-called "own" polarization of grating monochromator must be taken into account when calibrating the instrument. The light of the filament lamp used as a light source is depolarized, but due to "own" polarization of the monochromator, the light intensity after polaroid is modulated with the frequency of polarization modulation and coherently with its vector direction. The depth of this modulation has an irregular spectral dependence and should be determined for spectral measurements. The problem has been solved by means of a scattering etalon - standard scatter covered by MgO. This covering is known to have an albedo close to $\xi = 1.0$ and no additional polarization is brought about in the process of scattering. This etalon can be inserted into the scattering volume of the chamber, and intensities of parallel to the scattering plane $I_{||}$ and perpendicular $I_{\perp}$ polarized components of scattered by etalon light in the whole spectral region can be registered. Both intensities are proportional to the source beam ones with the same coefficient when the polarization components $D_{||,\perp}$ are measured. The sum $S = I_{||} + I_{\perp}$ (or, more correct, the inverse value S^{-1}) gives the throughout spectral dependence of the instrument sensitivity.

The instrument calibration in km^{-1} sr^{-1} units was carried out by means of the comparison of the spectropolarimeter data with simultaneous measurements with the serial aerosol nephelometer FAN. This instrument has a certificate calibration. The correlation coefficient R between both instrument data (about 50 realizations in the range D = 0.03 - 0.3 km^{-1} sr^{-1}) and standard deviation from regression line

ΔD were found to be equal R = 0.993 and ΔD = ± 5%. Both instruments are supplied by airflow heaters, allowing the characteristics of light scattering to be obtained by "dry" aerosol and to estimate the condensation activity of aerosol particles.

The iterative technique used for the solution of the equations connecting the polarization characteristics and aerosol size distribution in discrete form is described in Isakov (1997, 1998). The grid includes 45 kernel functions (nine values of n and five of χ). The range of radii r = 0.07 µm to 2.0 µm was divided into twenty size intervals. The mesh width for the real part of the refractive index was 0.03 and 0.01 for the imaginary part. The differences between measured and recalculated for retrieved size distributions optical parameters were usually within 3% to 10%. The results of inverse problem solution are presented for volume distributions dV(r)/dr. It must be noted that the kernel function sensitivity sharply decreases for r <0.07 µm and r>2.0 µm, so in these size regions, we can obtain only rough estimates of dV(r)/dr.

The daytime measurements by spectropolarimeter were started in February and finished in March 1999 (01.02.99 - 05.03.99). The common period of measurements was from 10 h to 23 h (local Moscow time). From the total number of records, about 200 were selected for the following processing (the rest were excluded since fluctuations caused by space - temporary inhomogeneity of the atmospheric aerosol and measurement defects). For these realizations, the inverse problem was solved - real and imaginary parts of the particle matter refractive index and aerosol volume size distributions in size range r = 0.03 µm to 2.0 µm were retrieved.

Some preliminary remarks must be addressed. 1) The time-averaged characteristics of atmospheric aerosol are well known to have seasonal and annual trends. Therefore, one month is too short a period to obtain reliable statistical estimations of aerosol parameters, and it is necessary to compare obtained parameters with ones for another measurements cycles. 2) In the measurements process, the instrumental errors for the spectropolarimeter (~1-5% registration noises, 2% to 3% spectral calibration and so on) in general account only for the lesser part of the total measurement errors, and the inhomogeneity of the temporary - space aerosol content accounts for the main part of them. These errors may be about 20% or larger. So the registration scheme was the following - spectral dependencies were twice registered (both for "wet" and "dry" aerosol). If the difference between two sequential measurements was greater than 15%, these records were excluded from total ensemble. 3) The spectropolarimeter was mounted in a laboratory room and in winter, the aerosol was heated when sampled even without a heater. The additional heating was about 2°C to 3°C when the external temperature T_{ext} was about zero and reached 10°C when T_{ext} ~ -20°C. Hence, in winter, we deal with a strongly drained aerosol even without using a heater.

The scattering coefficient $\boldsymbol{\sigma}$ is one of basic optical characteristic of atmospheric aerosol and is used as an input parameter in aerosol optical models. The value of D_{11} (45°, 0.55) is closely connected with σ (0.55) and can be used instead of σ.

From nephelometer data, the hour and daily averaged values and probability histogram of M were calculated. It was found that the level of dry aerosol content during the measurement period was close to the mean for February, i.e., measurement conditions were normal from the point of view of surface aerosol loading. The hour time series of M value and probability histogram are presented in Figures 1 and 2 correspondingly.

For parameterization of spectral dependencies of the extinction ε and scattering σ coefficients, the so-called Angstrom formula $\varepsilon,\sigma \sim\lambda^{-\alpha}$ is often used. An example of such approximation for D_{11} (45°) is shown in Figure 3. We have carried out the analysis of the applicability of this formula for all the realizations. It was found that for the majority or realizations (> 90%) in spectral region λ = 0.4 μm to 0.65 μm power law approximation is valid. The standard errors of it are about 2% to 5%; i.e., they are roughly equal to mean instrumental errors. In region λ > 0.65 the measured values of D_{11} are larger than calculated and the differences increase with wavelength. This effect increases with scattering coefficient σ decreasing and is connected with growth of the relative role of coarse aerosol in red spectral region. The parameter α varies from 0.5 in dense hazes up to 2.5 in very transparent atmosphere conditions and for total data ensemble the good correlation connection between α and σ exists (the correlation coefficient of the linear regression for logarithms α and σ R > 0.7. Such high correlation connection describes a half of the total α variance and may be used for retrieving α from an expression $\alpha = \text{const}\ \sigma^{\beta}$. The estimations of β value β = 0.45 and the standard deviation of α $\Delta\alpha = \pm$ 0.3 were obtained.

The second statistical ensemble was formed for data, obtained during November to December 1999 and January 2000. The number of measurements was about 230. The main statistical parameters for both ensembles were found to be close. Therefore, the correlation coefficient was R = 0.75, β = 0.43, $\Delta\alpha \approx$ 0.3. All of the items mentioned above allows considering this result as some stable statistical low.

For all realizations, the inverse problem was solved. As mentioned above, the results were presented in the form of aerosol volume distributions. We consider that this form is more suitable, because volume representation makes the aerosol fraction structure more apparent.

The variations of the particles' refractive index and total volume are presented in Figure 4. On the X-axis, the number of the measurement is plotted. Such presentation is used because they were only daytime measurements. Two periods are interesting - from 110 to 140 and from 180 up to 200. Both periods correspond to the atmospheric fronts passing - first corresponds to cool front 24.02 (see Figure 2), the second to warm (04.03). After the first front, the relative long interval (up to 28.02) of the accumulation of particle matter appeared, the total volume increased, and synchronously refractive index decreased. The second short period was connected with advective changing of the air mass, but the same behavior of n and V connection was observed.

The correlation diagram for ln(n) and lnV is presented in Figure 5. It follows from the diagram that an appreciable correlation connection exists between these values. The correlation coefficient is equal to R = 0.5; i.e., on the average, the particle chemical composition changes so that the average refractive index decreases with a total volume growth and particles accumulate on matter with a smaller n.

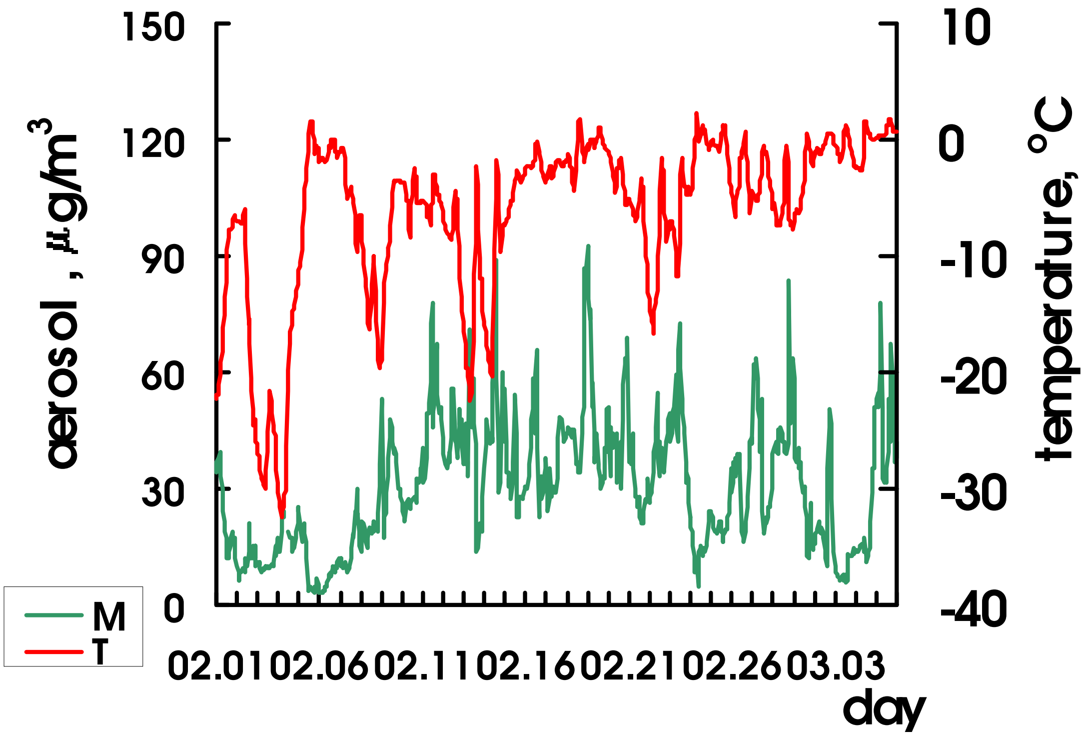

Figure 1. The time series of the hour values of submicron aerosol mass M(1). 2 - Air temperature.

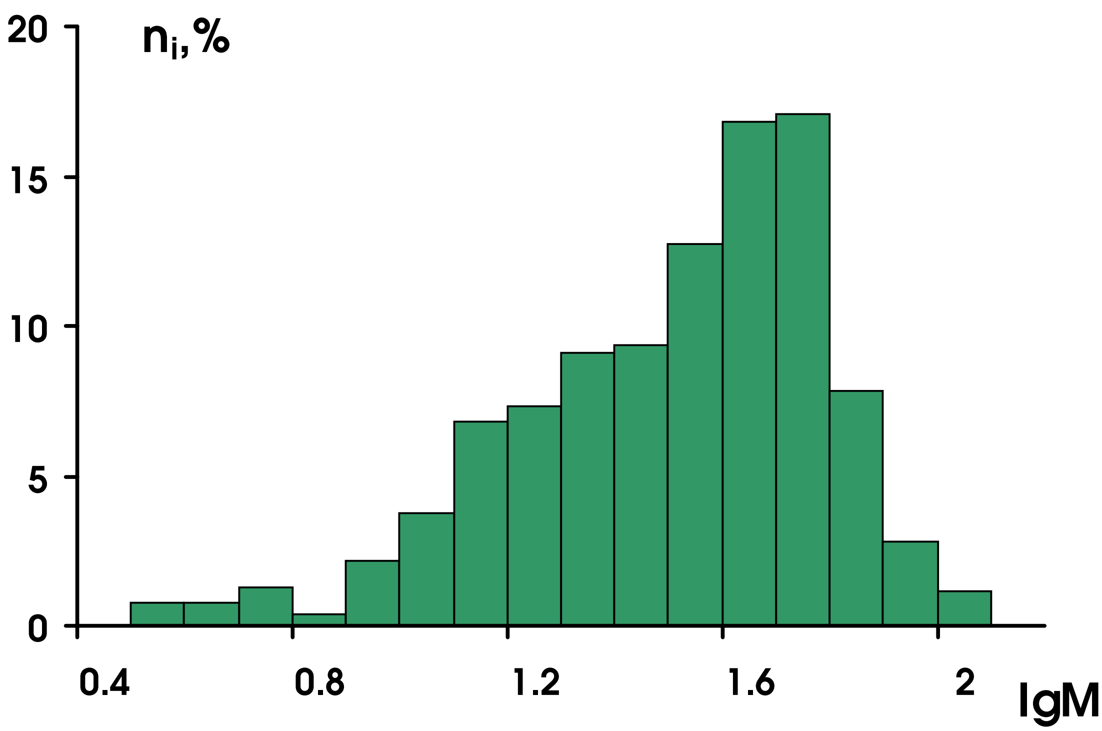

Figure 2. The frequency histogram of the logarithm of submicron aerosol mass values.

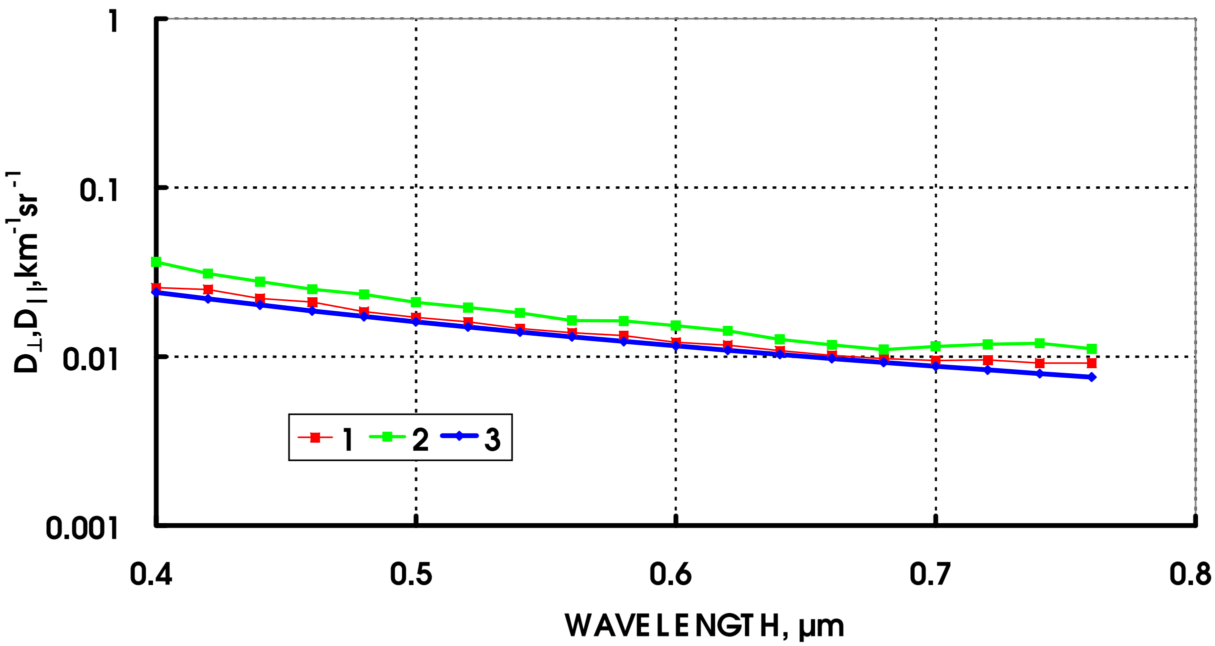

Figure 3. Examples of spectral dependencies of polarization components D (1,2). 3 - Angstrom line with $\alpha = 1.8$.

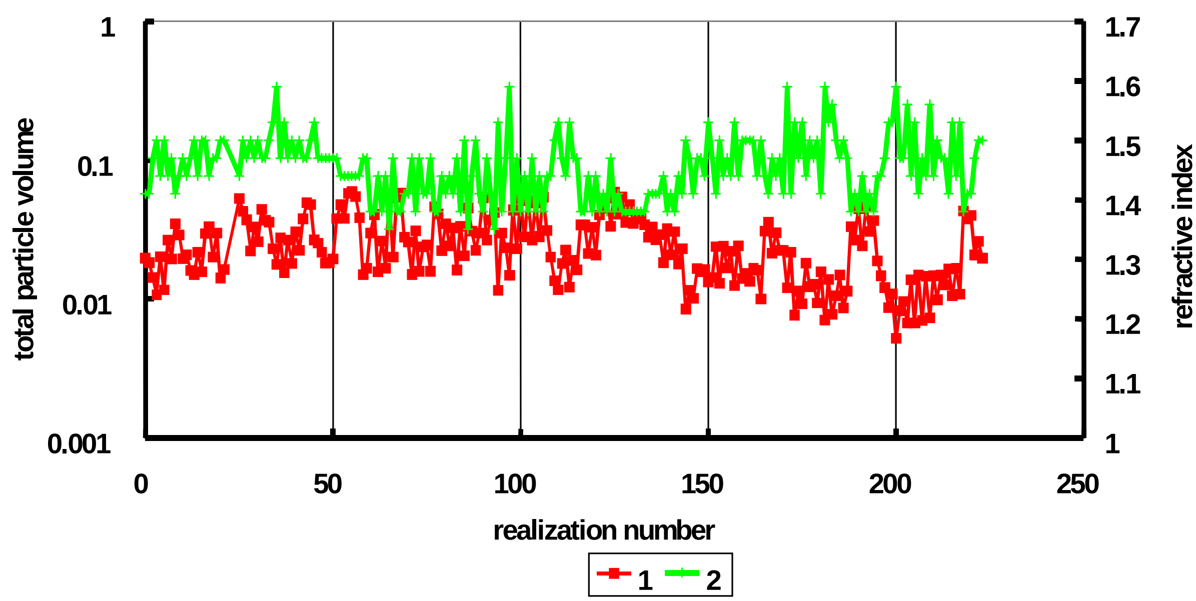

Figure 4. The time variations of total volume (1) and refractive index (2) of aerosol particles.

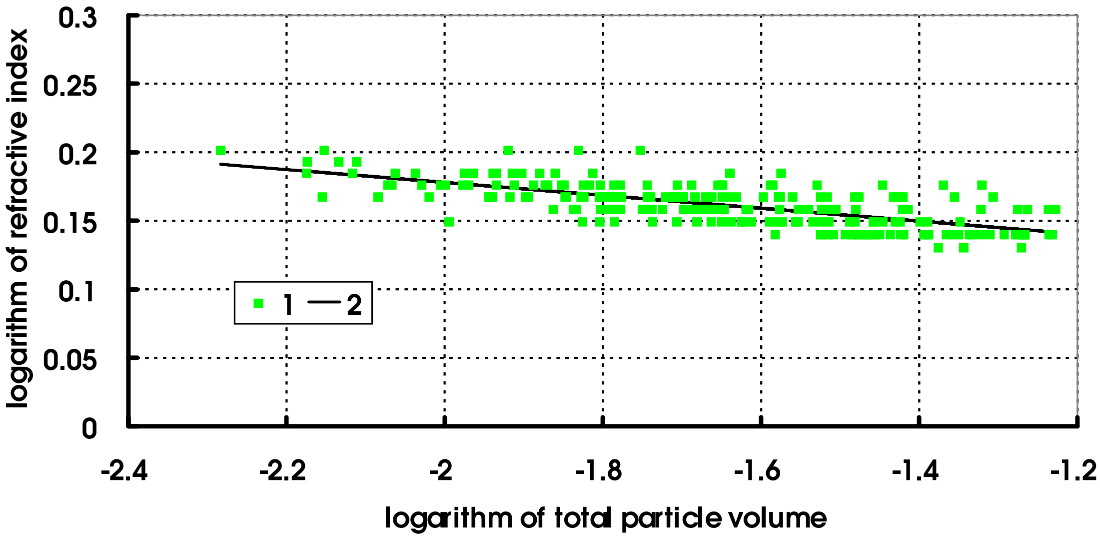

Figure 5. The correlation connection between total particle volume and refractive index (1), 2 - regression line.

Conclusions

1. The spectral dependencies of directed scattering coefficient D (45°, λ) in spectral region λ = 0.4 µm to 0.65 µm are well described by Angstrom's formula. A good correlation exists between Angstrom exponent α and scattering coefficient σ (correlation coefficient R ≈ 0.7), and this relationship may be expressed as $\alpha \sim \sigma^{-0.45}$. The standard deviation of the estimation of α is about 0.3.

2. The process of the particle matter accumulation is accompanied by the decrease of the particle refractive index. Commonly the refractive index value sharply increases when a cool atmospheric front is passing and sharply decreases with a warm one passing**.**

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References

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