Comparison of aerosol optical and microphysical characteristics in a local volume and on a long path

M.V. Panchenko, M.A. Sviridenkov,[1] A.S. Emilenko,[1] V.S. Kozlov, S.A. Terpugova, V.N. Uzhegov, and T.A. Dokukina

Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences, Tomsk
[1]Obukhov Institute of Atmospheric Physics, Russian Academy of Sciences, Moscow

Received January 23, 2007

The results of parallel measurements of light extinction on a horizontal path and light scattering in a local volume are considered. The inverse problem of light scattering is solved using the data of path and nephelometric measurements. In the most cases, an agreement between the aerosol particle size spectra is obtained for particle radii ranging from 0.1 to 0.5 μm. It is shown that the data obtained at the Aerosol Monitoring Station of IAO SB RAS are representative for the path located more than 500 m far from the station.

Introduction

Flow-through nephelometers with a closed scattering volume are widely used in the practice of optical studies of aerosol. Their doubtless advantage is the capability of performing round-the-clock measurements and controlled actions on aerosol. However, due to the limited wavelength range, as well as the set of scattering angles not including the aureole range, some design features of the air sampling path, and the small scattering volume, the measured optical characteristics are related, first of all, to the submicron aerosol fraction.

Measurements of spectral transparency of the atmosphere in the visible and infrared regions extend the range of the retrieved size spectrum. However, their use for solution of the inverse problem requires *a priori* knowledge of the complex refractive index.[1] This principle problem seriously restricts the possibilities of studying the particle size distribution, especially in the surface atmospheric layer, in which the diurnal behavior of relative humidity, significantly affecting the refractive index, is well pronounced.

The method of active spectral nephelometry developed by us[2] is capable, to some extent, of providing the necessary data on the refractive index and its diurnal dynamics under the specific conditions of the surface layer. At the same time, to introduce this method into the practice of everyday interpretation of path measurements, it was necessary to solve several technical problems.

The first of them concerns the comparison of optical characteristics measured in a local volume and on a long path. The second problem is the development of a technique for retrieving the refractive index under conditions of variable relative humidity of air and the corresponding application of data obtained to the inversion of data on the spectral aerosol extinction coefficients.

The three-week measurement cycle for solution of these technical problems was carried out in Tomsk in March–April, 2006 in cooperation between scientists of the Institute of Atmospheric Optics SB RAS and the Obukhov Institute of Atmospheric Physics RAS.

In this paper, we present the results of analysis of these observations and solution of the inverse problems for the data of nephelometric and path measurements.

Characterization of instrumentation, experimental conditions, and initial data

The instrumentation used in the experiment was installed at two points at the territory of Akademgorodok in Tomsk. The arrangement of measurement setups is shown schematically in Fig. 1.

The following parameters were determined:

1. The extinction coefficient $\varepsilon(\lambda)$ was measured by a multiwave path transparency meter (IAO) at a 830-m long horizontal near-ground path in the wavelength range 0.45–3.91 μm [Ref. 3]. Then the aerosol extinction coefficient $\beta(\lambda)$ was calculated from these data using the technique described in Ref. 4.

2. The directed scattering coefficient of the aerosol dry matter $\mu_0(\varphi)$ at an angle of 45° at wavelengths of 0.41, 0.51 and 0.63 μm and two orthogonal polarized components of scattered radiation at an angle of 90° at wavelengths of 0.45 and 0.52 μm. These parameters were measured by two FAN nephelometers, one of which (IAO) was set at the Aerosol Monitoring Station of IAO SB RAS, and another (IAP) was installed at an end of the path, at which transparency measurements were carried out. The distance between two measurement sites was ~ 650 m. In addition, the IAO nephelometer was equipped with a device for artificial humidification of the studied aerosol,[2] and the aerosol scattering parameters were measured at different values of the relative humidity in the range from 20–30 to 90%.

0235-6880/07/06 451-06 $02.00

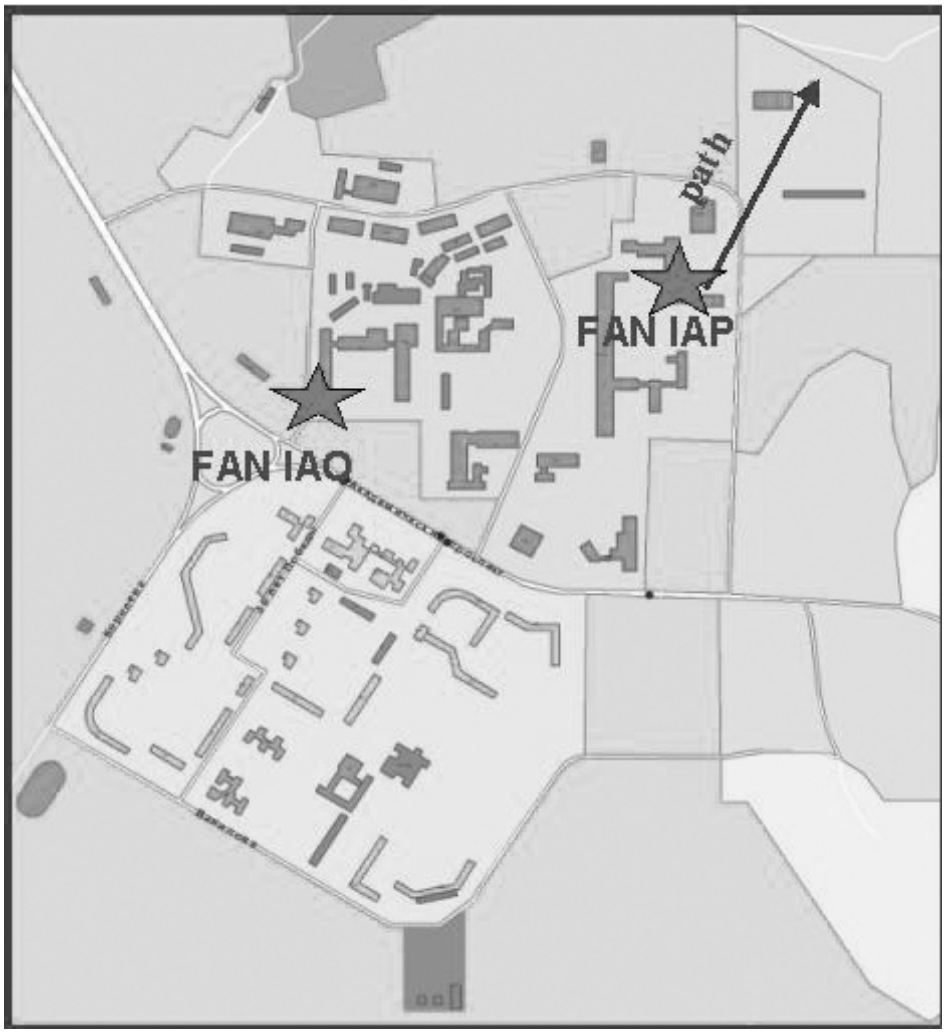

Fig. 1. Arrangement of measurement setups (asterisks) at the territory of Akademgorodok (Tomsk).

3. The mass concentration of black carbon M_{BC} was determined with an aethalometer[5] (IAO) and using the technique of sampling on filters (IAP).

Parallel measurements of parameters of light scattering by aerosol dry matter of and the mass concentration of black carbon at two sites made have allowed us to estimate the representativeness of the data obtained at the IAO Aerosol Monitoring Station for a path.

Meteorological data (temperature T, relative humidity RH, wind velocity and direction) obtained at the Tomsk station of Russian Hydrometeorological Service (http://www.gismeteo.ru/weather/ synarc.htm?29430) were used in analysis.

Estimation of the refractive index under actual atmospheric conditions

The inverse problem was solved for the optical parameters (the spectral extinction coefficients measured on the path and seven parameters of light scattering obtained with the FAN nephelometers) using the technique based on the Twitty algorithm[6] and adopted to inversion of particular parameters measured in the experiment considered.[2] The refractive index of aerosol matter (from FAN data) and the particle size distribution functions were determined from solution of the inverse problems. Analysis of the sensitivity of the measured parameters to particles of different size[2,7] has shown that nephelometric measurements allow one to determine reliably the size distribution function in the range $r = 0.05–0.6$ μm. When inverting the path measurements of transparency in the range 0.45–2.17 μm, it becomes possible to retrieve the particle spectrum in the wider size range.

However, measurements of the spectral transparency do not allow the refractive index of aerosol matter to be determined. Solution of the inverse problem in this case is possible only if it is set *a priori*. In our case, n was determined using the nephelometer data including angular and polarization scattering characteristics. This was performed both

for the dry matter (at RH ~ 20–30%) and at RH ~ 90% (the complete set of measurements at all wavelength was realized only at these values of humidity, and only the directed scattering coefficient $\mu_0(45°, 0.51\ \mu m)$ was recorded in the short range). It should be noted here that, as our previous research has shown, the set of the measured parameters and the applied technique for inversion do not allow separate determination of the real and imaginary parts of the refractive index, but only their linear combination.[2] Thus, the optical constants and the size spectrum were determined with aerosol assumed to be non-absorbing. However, it is not principally significant for this work. Virtually the same particle size distribution (accurate to errors of retrieval) corresponds to different values of the refractive and absorption indices with the difference between them kept the same.[2] Consequently, all the conclusions drawn from the comparison of the size spectra obtained from the results of inversion of the nephelometric and path measurements are also true if the aerosol absorption is taken into account.

It was also assumed that the refractive index has no spectral dependence within the visible and near IR wavelength regions. According to Ref. 8, it is true for the main components of atmospheric aerosol.

Then the refractive index was estimated *in situ*, i.e., at the relative humidity of air observed in the atmosphere at the time of measurement of the spectral aerosol scattering and extinction coefficients.

For this purpose, the volume concentration of submicron aerosol at the initial humidity and RH = 90% were calculated through integration of the volume particle size distributions dV/dr obtained from the solution of the corresponding inverse problem. Then it was checked how the change in the refractive index corresponds to the increment of the aerosol volume in the condensation process, i.e.,

$$n_{a,90} = (n_{a,0}V_{a,0} + n_w V_w)/V_{a,90}, \qquad (1)$$

where $n_{a,0}$ is the refractive index of aerosol matter and $V_{a,0}$ is the volume concentration of aerosol at the initial humidity; $n_{a,90}$ is the refractive index of aerosol matter and $V_{a,90}$ is the volume concentration of aerosol at a humidity of 90%; $n_w = 1.33$ is the refractive index of liquid water; V_w is the volume of condensed water (i.e., increment of the volume of aerosol particles, $V_w = V_{a,90} - V_{a,0}$). The error in determination of the refractive index for our data set and for the applied technique for solution of the inverse problem depends on the error of measurement of the light scattering characteristics, which, in their turn, are different for different values of the directed scattering coefficient $\mu(45°, 0.51\ \mu m)$. At $\mu = 20\ Mm^{-1}\cdot sr^{-1}$ the error of reconstruction of n amounts to ~ 0.01 and increases to ~ 0.08 at $\mu = 1\ Mm^{-1}\cdot sr^{-1}$ [Refs. 7 and 9]. When analyzing the experimental results, we rejected the data for which the deviation of the refractive index $n_{a,90}$ calculated by Eq. (1) from the value obtained from solution of the inverse problem exceeded the expected error. It should be said that, for the data, the optical discrepancy at whose inversion did not exceed 5%, Equation (1) was usually fulfilled except a few cases.

The change in the volume of aerosol particles was approximated by the Hanel formula[10]:

$$V_{a,RH} = V_{a,0}(1 - RH/100)^{-\chi}, \qquad (2)$$

where RH is the current value of relative humidity. The parameter χ was calculated using the values of the volume concentration of aerosol particles at the initial relative humidity and at RH = 90%:

$$\chi = -\frac{\ln V_{a,90} - \ln V_{a,0}}{0.1}. \qquad (3)$$

The value of the refractive index was calculated as

$$n_{is} = \frac{n_{a,0}V_{a,0} + 1.33(V_{a,is} - V_{a,0})}{V_{a,is}}, \qquad (4)$$

where the subscript "is" corresponds to the relative humidity in the *in situ* atmosphere. The value of n_{is} calculated in this way was used for inversion of the path data.

The range of variations of the refractive index of the aerosol dry fraction in the considered experiment was 1.46–1.62 (mean value of 1.57±0.04), *in situ* it was 1.42–1.57 (mean value of 1.50±0.04).

Results of investigations

Mean, maximum, and minimum values of the measured parameters and their rms deviations recorded during the experiment are summarized in Table 1. For comparison, we present the mean values and ranges of variations of some aerosol parameters in 2005: $\mu_0(45°, 0.51\ \mu m) = (11.32 \pm 9.65)\ Mm^{-1}\cdot sr^{-1}$ (0.22–108), $M_{BC} = (1.55 \pm 1.22)\ \mu g/m^3$ (0.11–12.1), $\beta(0.5\ \mu m) = (270 \pm 120)\ Mm^{-1}$ (42–1200). Thus, the ranges of variations and the rms deviations of the parameters observed during the experiment are only 1.5–2 times lower than those observed during the entire previous year. This fact suggests that the obtained experimental data are sufficiently representative and statistically provided.

Table 1. Mean values, rms deviations, and ranges of variations of the measured parameters

Parameter	Mean	rmsd	Min	Max
$\mu_0(45°, \lambda = 0.51\ \mu m)$, $Mm^{-1}\cdot sr^{-1}$	7.58	4.82	0.34	41.15
$\beta(0.5\ \mu m)$, Mm^{-1}	212	96	15	676
M_{BC}, $\mu g/m^3$	1.24	0.90	0.048	10.3
T, °C	–3.2	6.4	–22.0	11.0
RH, %	77.4	20.0	37	100

The correlation coefficients between different measured parameters are shown in Table 2.

Table 2. Correlation coefficients between parameters measured at different points

Parameter 1	Parameter 2	Correlation coefficient	Number of measurements
μ_0(45°, 0.41 μm), IAO	μ_0(45°, 0.41 μm), IAP	0.97	65
μ_0(90° ⊥, 0.41 μm), IAO	μ_0(90° ⊥, 0.41 μm), IAP	0.96	65
μ_0(90° ∥, 0.41 μm), IAO	μ_0(90° ∥, 0.41 μm), IAP	0.95	65
μ_0(45°, 0.51 μm), IAO	μ_0(45°, 0.51 μm), IAP	0.97	65
μ_0(90° ⊥, 0.51 μm), IAO	μ_0(90° ⊥, 0.51 μm), IAP	0.96	65
μ_0(90° ∥, 0.51 μm), IAO	μ_0(90° ∥, 0.51 μm), IAP	0.97	65
μ_0(45°, 0.63 μm), IAO	μ_0(45°, 0.63 μm), IAP	0.96	65
μ_0(45°, 0.51 μm), IAP	β(0.45 μm)	0.68	63
μ_0(45°, 0.51 μm), IAO	β(0.45 μm)	0.62	89
μ_0(45°, 0.51 μm), IAP	β(0.45 μm) − β(1.22 μm)	0.72	63
M_{BC}, aethalometer, IAO	M_{BC}, filters, IAP	0.88	23

It follows from Table 2 that the high correlation was observed between the values of the directed scattering coefficient of the aerosol dry matter measured with the nephelometers despite the two measurement sites are spatially separated. The values of the mass concentration of black carbon obtained at the two measurement sites correlate at a level of $\cong 0.9$. It is an evidence of the quite good spatial homogeneity of the field of the submicron aerosol fraction.

At the same time, the correlation between the scattering and the aerosol extinction in the path measurements is significantly lower. It may be caused by several reasons.

The *first* reason is that the measurement of seven parameters of the scattered radiation with the nephelometer takes only 4–5 minutes, while the measurement of the spectrum of the extinction coefficient in the wavelength range 0.45–3.91 μm requires ~ 25–30 minutes. The aerosol concentration and, consequently, the components of the scattered radiation can change several times during this time.

Some examples of temporal variations of the signal μ_0(45°, $\lambda = 0.51$ μm) (in relative units) observed during the measurements by the nephelometer installed near the sensing path are shown in Fig. 2. It can be seen that the situations both with the high stability of the signal and with its significant variability are observed.

The *second* reason is the spatial variability of the aerosol extinction on the path. The signals recorded by a photodetector are averaged over the sensing path and, consequently, can differ from the signals, which would be observed directly near the nephelometer air sampler.

The *third* reason is the variability of the relative contribution of the coarse fraction to the extinction in the visible region. As follows from Table 2, the difference between the extinction coefficients at wavelengths of 0.45 and 1.22 μm, which can be considered as the extinction coefficient of the submicron aerosol fraction, a few better correlates with the scattering coefficient than the extinction coefficient at a wavelength of 1.22 μm does.

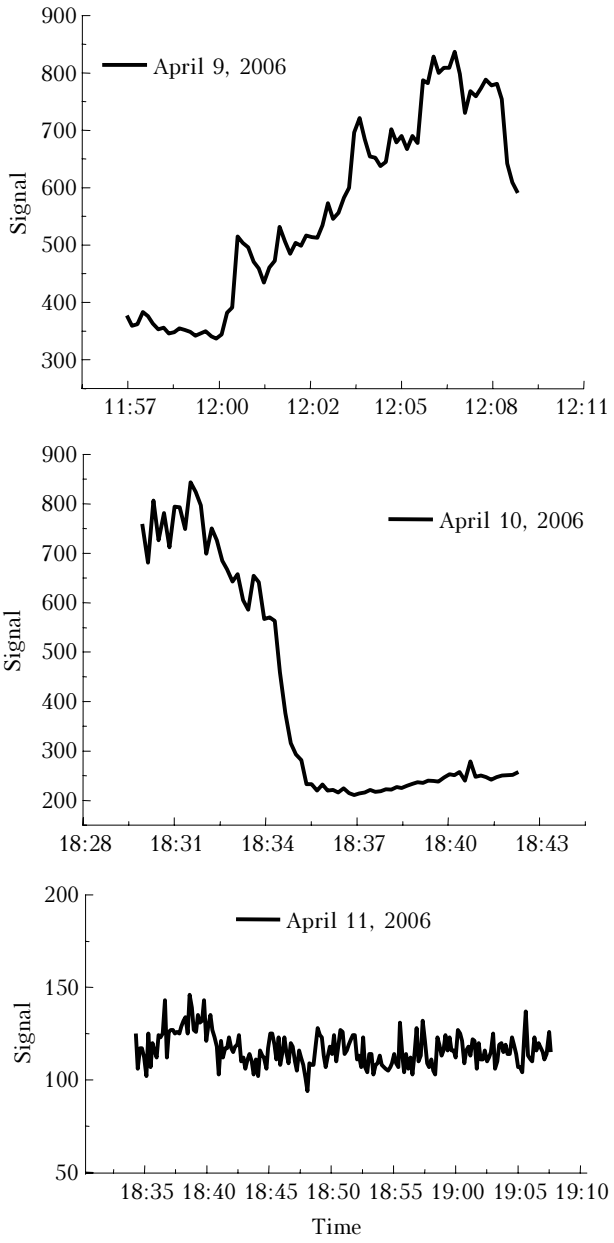

Fig. 2. Temporal variability of scattering near the path.

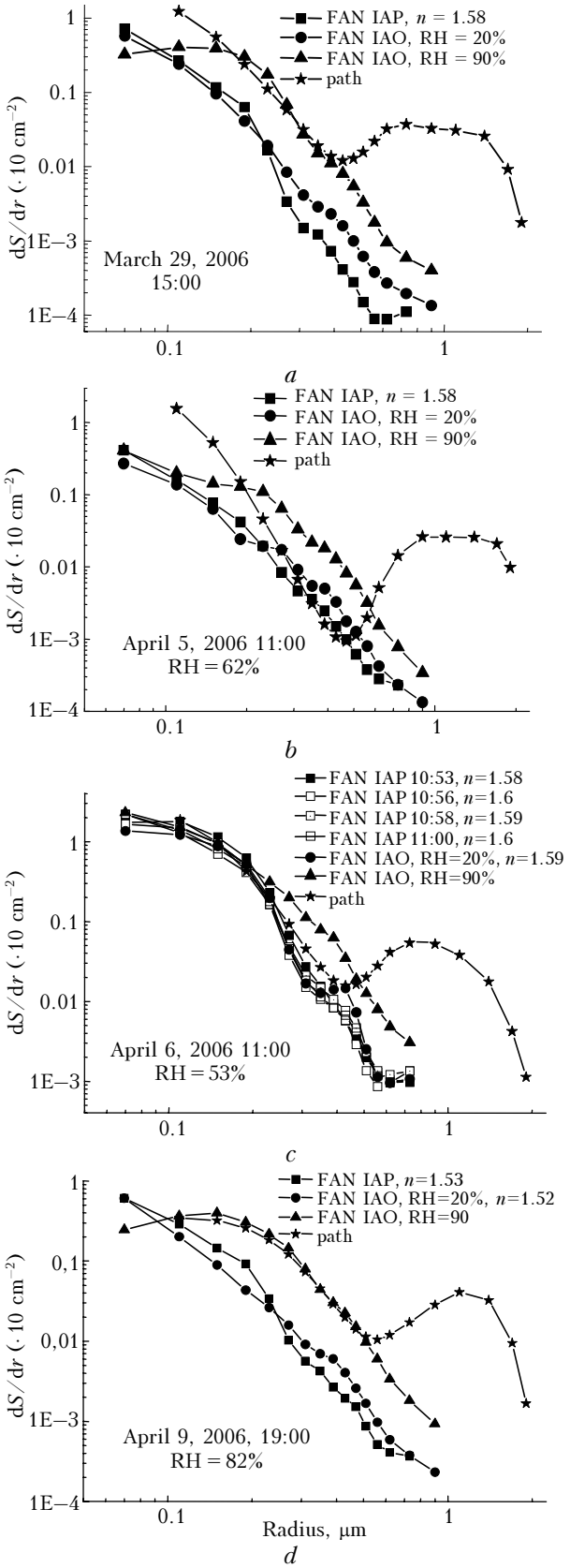

Fig. 3. Particle size distribution functions and the refractive index retrieved from the solution of the inverse problem for the measurements with the nephelometers and the path transparency meter.

In investigations of atmospheric aerosol, because of the large size range of aerosol particles (from hundredth to tens of micrometers), the selection of measurement instruments and methods adequate to a specific size range and the estimation of the boundaries of this range for every method are of principal significance. Theoretical analysis and numerical simulation partially remove this problem. Nevertheless, they also need in confirmation by data of field measurements. The comparison of the results of solution of the inverse problem for the path and local measurements (Fig. 3) has shown that the size spectra retrieved from the path measurements usually exceed those obtained from the inversion of the nephelometric data for the dry matter.

The particle size distributions and the values of the refractive index obtained from the data of the two nephelometers for the dry fraction coincide quite well in the size range where the measured parameters allow the reliable reconstruction of the microstructural parameters.

In many cases, the distributions obtained from the inversion of the data on the aerosol extinction coefficient on the path are close to the distributions retrieved from measurements at the Aerosol Monitoring Station at the relative humidity RH = 90% in the size range $r = 0.1$–0.5 μm. It is true at least for the situations when the relative humidity in the atmosphere was higher than 75% (see Figs. 3*a*,*d*). Significant differences can be observed in the distributions at the lower humidity (Figs. 3*b*,*c*) due to the discussed spatiotemporal inhomogeneity of the aerosol optical parameters along the path.

Thus, it follows from the data presented that, at the known refractive index, the results of measurements of the spectral transparency in the visible and IR regions allow the reliable reconstruction of the size spectrum up to $r \cong 1$ μm. Formally, the solution of the inverse problem can also yield the spectrum of the medium fraction (see Fig. 3). At the same time, it should be noted that the correctness of the distribution function for the medium fraction depends on how close is the refractive index for the medium fraction that obtained for submicron particles.

Conclusions

The comparison of the measurements of the light scattering characteristics and the black carbon at the IAO Aerosol Monitoring Station and near the horizontal path, as well as the results of solution of the inverse problem, shows the high correlation between the parameters considered at the two measurement sites. Therefore, the data of the Aerosol Monitoring Station can be considered as representative for the path lying more than 500 m far from it.

The size spectra retrieved from the spectral polarimetric measurements in a local volume and the measurements of the spectral transparency on a horizontal path are in satisfactory agreement in the range from 0.1–0.15 to 0.4–0.5 μm. The results

obtained are tentative and require confirmation with more extended experimental data.

Acknowledgements

The work was supported in part by the Russian Foundation for Basic Research (Grants No. 06-05-64393 and 07-05-00860).

References

1. M.A. Sviridenkov, Atmos. Oceanic Opt. **14**, No. 12, 1022–1025 (2001).
2. M.V. Panchenko, M.A. Sviridenkov, S.A. Terpugova, and V.S. Kozlov, Atmos. Oceanic Opt. **17**, Nos. 5–6, 378–386 (2004).
3. Yu.A. Pkhalagov, V.N. Uzhegov, and N.N. Shchelkanov, Atmos. Oceanic Opt. **5**, No. 6, 423–426 (1992).
4. Yu.A. Pkhalagov and V.N. Uzhegov, Opt. Atm. **1**, No. 10, 3–11 (1988).
5. V.S. Kozlov, V.V. Burkov, M.V. Panchenko, and V.P. Shmargunov, in: *Science for Industry* (2003), pp. 15–19.
6. J.T. Twitty, J. Atmos. Sci. **32**, 584–591 (1975).
7. M.V. Panchenko, M.A. Sviridenkov, and S.A. Terpugova, Proc. SPIE **5397**, 43–49 (2003).
8. V.E. Zuev and G.N. Krekov, *Optical Models of the Atmosphere* (Gidrometeoizdat, Leningrad, 1986), 256 pp.
9. S.A. Terpugova and M.V. Panchenko, in: *Abstracts of Reports at International Symposium of CIS "Atmospheric Radiation,"* St. Petersburg (2004), pp. 82–83.
10. G. Hanel, Adv. in Geophys. **19**, 73–188 (1976).