METHOD FOR DETERMINATION OF LIGHT BACKSCATTERING AND EXTINCTION COEFFICIENTS IN TROPOSPHERIC AEROSOL LAYERS USING ELASTIC– AND RAMAN–BACKSCATTER–BASED LIDAR

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The ratio of lidar return at unbiased wavelength of 532 nm to a sum of lidar returns from two regions of the rotational spectrum of Raman scattering (RS) by nitrogen and oxygen molecules is used to measure the light backscattering coefficients. The ratio of the rotational RS intensities from two spectral regions can determine the vertical temperature profile; the ratio between the RS signals and temperature values from two adjacent segments of the path are used to calculate the extinction coefficients. The measurement errors and the ways to lower them are also discussed in the paper.

INTRODUCTION

The problem of inverting the lidar equation with respect to the backscattering and extinction coefficients gave rise to many publications where some additional assumptions without which the equation could not be solved were discussed. The problem is that the relation between aerosol backscattering and extinction coefficients

$$\gamma(h) = \beta(h) / \alpha(h) , \tag{1}$$

referred to as the lidar ratio, depends on microstructure of aerosols and due to their variety is a strongly variable value.

During the last several years in sounding the stratosphere and upper layers of the troposphere generally recognized is the method of determining the scattering as

$$R(h) = 1 + \beta^{a}(h) / \beta^{m}(h) , \tag{2}$$

where β^{a} and β^{m} are the backscattering coefficients of aerosol and molecular components of the atmosphere, respectively.

The method consists in the lidar response calibration using the return from the height h_c where the values $R(h_c)$ can be assigned *a priori* based on the known information about optical properties of the atmosphere. This calibration gives the following expression for the function $R(h)$:

$$R(h) = R(h_c)\,\frac{S(h)\,\beta^{m}(h_c)\exp\left\{-2\int\limits_0^{h_c}\alpha(x)\,\mathrm{d}\,x\right\}}{S(h_c)\,\beta^{m}(h)\exp\left\{-2\int\limits_0^{h}\alpha(x)\,\mathrm{d}\,x\right\}} , \tag{3}$$

where $S(h)$ is the lidar signal multiplied by the height squared; $\alpha(h) = \alpha^{a}(h) + \alpha^{m}(h)$ is the sum of the coefficients of light extinction by aerosol and molecular components of the atmosphere.

The profile of molecular scattering $\beta^{m}(h)$ can be assigned using the available models of molecular atmosphere or, that is more specific, calculated from aerological data on pressure and temperature.

The errors of this method were analyzed in, e.g., Ref. 1. The known arbitrariness in the choice of h_c and $R(h_c)$ and other sources of errors apart, we can note that the method works well until

$$1 - \exp\left\{-2\int\limits_h^{h_c}\alpha^{a}(x)\,\mathrm{d}\,x\right\} \le |\delta| , \tag{4}$$

where δ is the allowable level of relative measurement error.

To put it differently, the error in assigning the lidar ratio (1) does not contribute a substantial error to Eq. (3) only with insignificant aerosol turbidities of the atmosphere when contribution of an aerosol component to the optical depth is of the order of one tenth of unit. In other situations, the error in assigning the lidar ratio which is introduced in either explicit form as, e.g., in Ref. 2 or in implicit form as in Klett formula,[3] plays an important role.

The lidar ratio does not enter in the Klett algorithm formally. But this is the result of those assumptions which are made in deriving the formula of, e.g., power dependence between the backscattering and extinction coefficients and, in the simplest case, a linear dependence. The key advantage of the algorithm consists in the use of reference value in the remote segments of the path that allows one to avoid the solution instability due to the increased errors but does not exclude the possibility of substantial errors if the real behavior of the lidar ratio differs strongly from the assumptions taken in deriving the formula.

Integration of the lidar equation without any assumptions on the type of the function (1) gives the following solution[4]:

$$\alpha(h) = S(h) \Bigg/ \left\{ \frac{\alpha(h_c)}{S(h_c)} - 2 \int\limits_{h_c}^{h} S(x)\, \mathrm{d}\, x + \frac{S(h_c)}{\gamma(h_c)} \int\limits_{h_c}^{h} \gamma'_x(x) \exp\left[-2 \int\limits_{h_c}^{x} \alpha(\xi)\, \mathrm{d}x \right] \mathrm{d}\, x \right\}, \qquad (5)$$

where $\gamma'_x(x)$ is the derivative of the lidar ratio with respect to distance. The remaining designations were accepted above.

The formula gives the solution for $\alpha(h)$ in an implicit form, if the function $\gamma'_x(x)$ is assigned. In Ref. 5, there is an example of iteration algorithm for calculating the extinction profile along the vertical path using formula (5). In this case, it was assumed that variations in $\gamma(h)$ occurs due to backscattering redistribution between molecular and aerosol components of the atmosphere. It was also assumed that the lidar ratio of aerosol component remains constant along the path. These assumptions results in dependence of $\gamma'_x(h)$ on $\alpha(h)$.

Formula (5) converts to the Klett formula if one assumes $\gamma'_h(h) = 0$ and reverses the positions of limits of integration in the second term of the denominator. This allows one to estimate the conditions of satisfactory operation of the Klett algorithm.

The analysis of errors of different methods of solving the lidar equation is presented in Ref. 6, and it is shown there that the method similar to the Klett algorithm operates satisfactorily when the condition

$$\gamma'_h(h) \,/\, \gamma(h) \ll \alpha(h)\ , \qquad (6)$$

holds, i.e., along sufficiently optically dense paths with a slowly varying lidar ratio. The same is evident from the numerical experiment[5] and the comparative analysis made by the authors of Ref. 7.

Thus, it can be noted that satisfactory solution of the lidar equation can not be found for the paths with sharply varying optical characteristics if we restrict our consideration to the lidar return reception only at frequency of laser transmitter.

An idea of calibration using molecular datum obtained by recording the returns at frequencies of Raman scattering (RS) by air molecules has long been put forward and realized experimentally.[8–11] This enables one to separate the problems of calculating the coefficients of backscattering and extinction. But because of small RS cross sections and more complicated instrumentation, this method has not received wide acceptance. Only in the early nineties the authors of Ref. 7 published the interesting results of studying the aerosols in the upper layers of troposphere using the method based on RS. The powerful laser operating in the UV ($\lambda = 308$ nm) where the RS cross sections are large made great contribution to this direction.

In the aforementioned method, there are two points which require *a priori* assumptions or the use of some extra measurements. First, since the RS is used at the wavelength of nitrogen vibrational line ($\lambda = 322$ nm), it is necessary to introduce the value equal to some power of wavelength ratio of elastic λ_0 and RS λ_R components of the return signal

$$(\lambda_0 \,/\, \lambda_R)^{\kappa}. \qquad (7)$$

This ratio takes into account the spectral behavior of the coefficient of light extinction by an aerosol component. The power index depends on the aerosol type that brings some uncertainty in the method. Second, since the temperature profile in the lidar experiment does not change, it must be either assigned using the atmospheric model or obtained from the aerological measurements.

Described below is the method in which the aforementioned facts are absent. The instrumentation for realization of this method is outlined, and the examples of its usage for determining the coefficients of backscattering and extinction in aerosol layers of troposphere are given.

THEORETICAL GROUNDS OF THE METHOD

The method described below consists in recording the backscattering signals at unbiased wavelength λ_0 and in the two narrow wavelength segments of the rotational RS spectrum of nitrogen and oxygen molecules.

Each of the three signals is described by the lidar equation

$$P_i(h)\, h^2 = K_i\, G(h)\, \beta_i(h) \exp\left\{ - \int\limits_0^h [\alpha_0(x) + \alpha_i(x)]\, \mathrm{d}\, x \right\}, \qquad (8)$$

where i is the number of spectral segment taken the values 0, 1, 2, ...; $P_i(h)$ is the signal power in the ith segment of the spectrum received from the distance h; K_i is the instrumental constant for the channel recording the radiation in the ith spectral segment; $G(h)$ is the geometric function taking account of overlapping the directional patterns of receiving and transmitting lidar antennas; and, β_i and α_i is the coefficients of backscattering and extinction, respectively. For the latter ones we can write the following relations:

For elastic scattering

$$\beta_0(h) = \beta_0^{\mathrm{a}}(h) + \beta_0^{\mathrm{m}}(h)\ , \qquad (9)$$

$$\alpha_0(h) = \alpha_0^{\mathrm{a}}(h) + \alpha_0^{\mathrm{m}}(h)\ , \qquad (10)$$

where the superscripts a and m stand for aerosol and molecular components, respectively. Moreover,

$$\beta_0^{\mathrm{m}}(h) = N(h)\, \mathrm{d}\sigma^{\mathrm{m}}/(\mathrm{d}\Omega)\ , \qquad (11)$$

where $N(h)$ is the molecular concentration at the altitude h; $\mathrm{d}\sigma^{\mathrm{m}}/(\mathrm{d}\Omega)$ is the differential cross section of Rayleigh backscattering for air molecules.

For rigid scattering

$$\beta_i(h) = N(h) \left[\eta\, \frac{\mathrm{d}\sigma_i^{\mathrm{N}_2}}{\mathrm{d}\Omega} + \mathrm{k}\, \frac{\mathrm{d}\sigma_i^{\mathrm{O}_2}}{\mathrm{d}\Omega} \right], \qquad (12)$$

where η, κ is the relative content of nitrogen and oxygen in air; $\mathrm{d}\sigma_i^{\mathrm{N}_2}/(\mathrm{d}\Omega)$ and $\mathrm{d}\sigma_i^{\mathrm{O}_2}/(\mathrm{d}\Omega)$ have the meaning of Raman backscattering cross sections averaged over rotational states falling into the specific intervals of rotational spectrum.

$$\alpha_i(h) = \alpha_i^{\mathrm{a}}(h) + \alpha_i^{\mathrm{m}}(h)\ .$$

Since the maximum spectral shift is 32 Å at $\lambda = 5320$ Å, then

$$\alpha_0^{\mathrm{m}}(h)/\alpha_2^{\mathrm{m}}(h) = (\lambda_2/\lambda_0)^4 = 1.024\ . \qquad (13)$$

The spectral behavior of the aerosol extinction coefficient is pronounced weakly, and the difference in the extinction coefficients at λ_0 and λ_2 will be observed only in the third symbol after the point. Therefore during the sounding of aerosol formations comparable in optical density with a molecular component of the atmosphere or, all the more, exceeding it substantially, with the error in optical depth no more than 1%, in the system of equations (8) for all *i* it is possible to assume that

$$\alpha_0(h) + \alpha_i(h) = 2\,\alpha_0(h) \tag{14}$$

and to obtain, taking account of Eqs. (9)–(12), the following relation

$$P_0(h)/[P_1(h) + P_2(h)] = K\,R(h)\ , \tag{15}$$

where $R(h)$ is the scattering ratio introduced by the formula (2), and the coefficient K is determined by the following expression:

$$K = K_0\,\frac{\mathrm{d}\sigma^{\mathrm{m}}}{\mathrm{d}\Omega}\left[K_1\left(\eta\,\frac{\mathrm{d}\sigma_1^{\mathrm{N_2}}}{\mathrm{d}\Omega} + \mathrm{k}\,\frac{\mathrm{d}\sigma_1^{\mathrm{O_2}}}{\mathrm{d}\Omega}\right) + K_2\left(\eta\,\frac{\mathrm{d}\sigma_2^{\mathrm{N_2}}}{\mathrm{d}\Omega} + \mathrm{k}\,\frac{\mathrm{d}\sigma_2^{\mathrm{O_2}}}{\mathrm{d}\Omega}\right)\right]^{-1}. \tag{16}$$

Strictly speaking, the coefficient K may depend on altitude due to temperature variations that, in its turn, results in redistribution of intensities between the rotational RS levels. But it is possible to show that with the appropriate choice of position and width of spectral intervals the temperature dependence of signals summation $P_1(h) + P_2(h)$ is in fact eliminated.[12,13] Let us introduce the designation

$$S(h) = [P_1(h) + P_2(h)]\,h^2\ . \tag{17}$$

To calculate the extinction coefficient in the segment Δh, we use the ratio of the sum of rotational RS signals at two adjacent sections of the path. Taking into account the relations (12) and (14) it is possible to obtain

$$\frac{S(h_{j+1})}{S(h_j)} = \frac{G(h_{j+1})N(h_{j+1})}{G(h_j)\ N(h_j)}\,\exp\left\{-2\int\limits_{h_j}^{h_{j+1}}\alpha(x)\,\mathrm{d}\,x\right\}. \tag{18}$$

As a consequence of the equation of ideal–gas state and barometric formula, we can write

$$\frac{N(h_{j+1})}{N(h_j)} = \frac{T(h_j)}{T(h_{j+1})}\,\exp\left\{-\frac{\mu\,g}{R}\int\limits_{h_j}^{h_{j+1}}\frac{\mathrm{d}h}{T(h)}\right\}, \tag{19}$$

where $T(h)$ is the air temperature at the altitude h; μ is the mass of one mole of air; R is the universal gas constant; and, g is the free–fall acceleration.

If we substitute Eq. (19) into Eq. (18), take the logarithm, and use the theorem of the mean and the formula of trapezoids for integral terms in a small interval of height $\Delta h_{(j,\,j+1)} = h_{j+1} - h_j$ then it is possible to derive the expression

$$\overline{\alpha}_{(j,\,j+1)} = \frac{1}{2\Delta\,h_{j,\,j+1}}\left[\ln\frac{G_{j+1}}{G_j} + \ln\frac{S_jT_j}{S_{j+1}T_{j+1}}\right] - \frac{\mu\,g}{4\,R}\,\frac{T_j + T_{j+1}}{T_j\,T_{j+1}}. \tag{20}$$

If the segment under study is distant so that the geometric function becomes constant then the first term in square brackets vanishes. As follows from formula (20), the profile of the extinction coefficient along the vertical path can be determined if the temperature profile is known.

The method of measuring the temperature from the rotational RS spectrum is presented, e.g., in Ref. 14, and as applied to the instrumentation used by the authors it is described in Refs. 15 and 16. The determination of the vertical temperature profile is based on optimal choice of such two segments of rotational spectrum for which the intensity ratio is most sensitive to temperature redistribution of rotational levels population. As shown in Ref. 14, at the temperature variation for one degree the variation of intensity ratio may be 1.5–2%.

The temperature profile is expressed through the signal ratio $L(h) = P_1(h)/P_2(h)$ by the formula

$$T(h) = a/[\ln L(h) - b]\ , \tag{21}$$

where a and b are the constants depending on choice of position and width of spectral segments.

The measured temperature profile enables one to solve Eq. (20) and calculate Rayleigh components of backscattering and extinction. To this end, it is sufficient to make use of formula (19), the known relation between molecular concentration and Rayleigh scattering, and the information about air pressure at zero or some other level which is taken as a reference one.

After the Rayleigh coefficient profiles of backscattering and volume scattering have been calculated, it is possible to calculate the backscattering coefficient of an aerosol component and to determine a portion of the latter one in the total extinction using Eqs. (2) and (15).

INSTRUMENTATION

The lidar used was described in Ref. 16 therefore we give only brief description here.

The laser transmitter emits pulses with 15–ns duration, energy per pulse of 50–80 mJ, and 10–Hz frequency at wavelength of 532 nm.

The transmitting antenna collimates a laser beam so that the radiation divergence equals one angular minute.

Relatively low energy characteristics of laser are partially compensated for by the return reception to the 2.2–m–diameter receiving antenna of lidar system of the laser sounding station.[17] The scattered radiation focused into a focal plane of the antenna is transmitted with a light–guide rope to a slit of double monochromator in which there are three spectral intervals whose centres correspond to the position of unbiased wavelength $\lambda_0 = 532$ nm and rotational lines of nitrogen with quantum numbers $J = 6$ and $J = 14$. Suppression of unbiased frequency background in the intervals $J = 6$ and $J = 14$ is no less than 10^8. The radiation in these three spectral intervals is recorded with photomultipliers FÉU–104 and pulse counters with a rate of 25 MHz.

MEASUREMENTS, CALIBRATIONS, AND ANALYSIS OF ERRORS

The measurements are carried out in the photon counting mode with signal storage over $5\cdot10^3$–10^4 laser pulses in sequence of time strobes whose duration corresponds to a 48–m spatial strobe. To decrease statistical fluctuations of signals, we make averaging over several (to ten) strobes so that spatial resolution can vary from 48 to 480 m.

Depicted in Fig. 1 is an example of realization of three signals described with the system of equations (8). The recorded signal is a mixture of legitimate signal, external noise of a detector, and noise of background illuminations. The mean noise component of signals is determined in each channel as the mean pulse counting rate in the strobes, the position of which at the time axis corresponds to distance of 40 km and larger. In processing the mean noise component is subtracted from the signal preliminary corrected for the noise of PM aftereffect.[19]

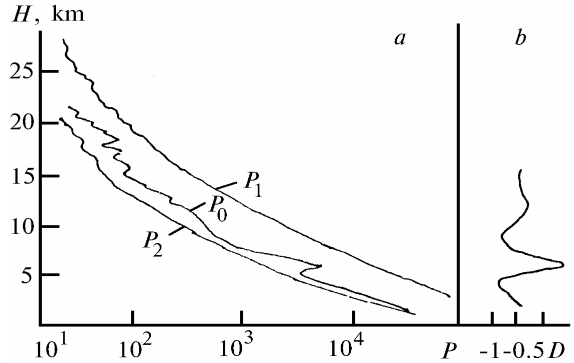

FIG. 1. Example of signal realization: at the unbiased wavelength (P_0) and in the spectral intervals of rotational RS (P_1 and P_2). The centres of intervals correspond to rotational quantum numbers of nitrogen molecules $J = 6$ and $J = 14$, respectively. P is the number of photons recorded in a time strobe corresponding to spatial distribution of 480 m. *The curve on the right is an example of choice of altitudes of calibration based on minimum signal ratio.* $D = \log [P_0 / (P_1 + P_2)]$.

DETERMINATION OF R–RATIO OF THE TOTAL BACKSCATTERING TO THE MOLECULAR SCATTERING

To determine $R(h)$ by the formula (15), it is necessary to know the coefficient K which is determined by the formula (16). The relations between the coefficients K_0, K_1, and K_2 from Eq. (16) are easily found experimentally, but the cross section of rotational levels are known with low accuracy. Therefore we use the known procedure of calibrating over the layer at some altitude h_c at which $R(h_c)$ can be assigned *a priori*. The altitudes, at which the signal ratio (15) is minimum, are chosen for calibration. For them the scattering ratio is assigned:

$$R(h_c) = 1 + \beta^{a}_{M}(h_c) / \beta^{m}_{M}(h_c) , \qquad (22)$$

where β^{a}_{M} and β^{m}_{M} are aerosol and molecular components of backscattering, respectively, which were borrowed from the model of Ref 18.

In the example represented in Fig. 1, the altitudes of 4.5 and 8 km can be taken as calibration ones. In accordance with the model, $R(h = 4.5\text{ km}) = 1.21$ and $R(h = 8\text{ km}) = 1.05$. If we use these values then the values 0.089 and 0.116 result for the coefficient K, i.e., a random spread of the calibration coefficient can be substantial. But in multiple iterations of calibration procedure, it becomes possible to estimate the mean and its variance. Our estimate over nine realizations in different days gave the result $K = 0.112 \pm 0.007$ that corresponds to a relative error of 6% and has been taken for estimating the measurement error in $R(h)$.

If the statistics of signals and noise is assumed to be Poisson, then it is possible to show that the relative error of measuring $R(h)$ does not exceed the value

$$\delta R \le \sqrt{(\delta K)^2 + 3(P + p_n) / 2(P - p_n)^2} , \qquad (23)$$

where K is the relative error of calibration previously assessed; P is the sum of signal and noise photocounts in the weakest signal; and, p_n is the estimate of the mean noise.

In the statistics usual in our measurements the relative error in determining $R(h)$ does not exceed 7% up to 8–km altitudes and gradually increases to 20% at the 15–km altitude.

DETERMINATION OF TEMPERATURE PROFILE

As mentioned above, the temperature is determined from the ratio of signals P_1 and P_2 in two segments of the rotational RS spectrum (formula (21)). The relative error in the temperature determination is expressed by the formula

$$\delta T \le \sqrt{(\delta a)^2 + T^2(\delta\alpha)^2 / a^2 + T^2 b^2(\delta b)^2 / a^2} . \qquad (24)$$

The empirical constants a and b are found using the procedure of minimization of sum of the squared deviations between the temperatures measured using sounding–balloon sensors and those calculated by formula (21). Calibration is made over a segment of the temperature profile at the 1.5–5 km altitudes where the relative fluctuations of signals are weak.

Figure 2 represents an example of superposition of the temperature profiles measured using the two methods. In the 1.5–5 km segment, the profiles coincide well within the limits caused by statistical fluctuations of signals and errors in the balloon measurements, and the possible errors do not exceed 10% of the errors caused by statistical fluctuations of signals. Therefore we take the estimate

$$\delta T(h) = T(h)\, \delta L(h) / a , \qquad (25)$$

for relative error in the temperature determination, or, if the photocount statistics is assumed to be Poisson, it is possible to write for the upper limit of relative error:

$$\delta T_j = 100 \frac{T_j}{a(P_j - p_j)} \sqrt{2(P_j + p_j)} , \%, \qquad (26)$$

where P_j and p_j are, respectively, the total photocount number and the mean number of noise pulses falling within the jth strobe. The T/a ratio is about 0.3. When the signal–to–noise ratio is 100 and there are 10^4 photocounts stored in the jth strobe, the relative error in the temperature determination is about 0.5% or about 1.5 K.

The following approximated formula

$$\mathrm{d}\bar{\alpha}_{j,\,j+1} = \tau^{-1}_{j,\,j+1} \sqrt{(\delta G_j)^2 + (\delta S_j)^2 + (\delta T)^2} \qquad (27)$$

can be written for relative error in determining the extinction coefficient using the algorithm (20). Here $\tau_{j,\,j+1}$ is the optical depth of the path segment $h_{j+1} - h_j$.

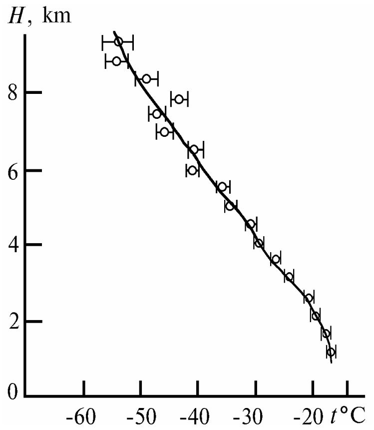

FIG. 2. *Example of superposition of temperature profiles measured with a sounding balloon (solid line) and a lidar for determining constants a and b entering in formula (21).*

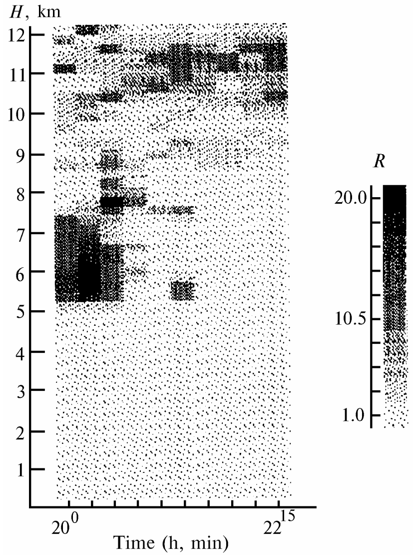

FIG. 3. *Variation of scattering ratio R in time.*

The first term in the radicand is of fundamental importance for the lidar used since the effect of geometric factor is appreciable to the 7–km altitude. The calculations by the formulas of geometrical optics provide only a rough estimate due to the complicated control over all parameters, on which the function of geometric factor depends. Therefore we have to estimate this function experimentally using the formula (20) applicable under conditions where the profile $\alpha(h)$ can be assigned based on the measured scattering ratio and model representations of aerosol. Such a case is depicted in Fig. 4, where a sample from a series of observations represented in Fig. 3 about vertical profiles of aerosol scattering made during 2.5 hrs is shown. There is a segment between 2 and 8 km in the profile where R has a stable small value so that the contribution of aerosol scattering is not substantial and can be estimated from model representations.

The accuracy of such an estimate depends on altitude, and it is 5–7% at the altitudes of 5 to 8 km. At altitudes below 5 km, the relative error in estimating the geometric factor increases to 10% and larger. This fact constrains the applicability of this method in the measurements using the aforementioned lidar, but it is not very important. At the altitudes where the action of the geometric factor reduces to the decrease of the signal inversely proportional to the squared distance, the first term in the radicand vanishes, and eventually the error in determining the extinction coefficient depends on statistical errors in the signal measurements. The multiplier $1/\tau$ in the formula (27) imposes rigid restrictions on these errors, if we speak about the sounding of a weakly turbid atmosphere or try to attain high spatial resolution.

In the existing lidar, the relative errors may be 15–30% in sounding aerosol formations with the scattering ratio $R > 10$ at altitudes of 5 to 8 km and spatial resolution of 0.5 km. Depicted in Fig. 4 *c* is the example of determining α using the described method (curve *1*) as compared to the calculation of α made with the assumption of existing the scattering by a cloud formation at an altitude of 6 km (Fig. 4 *a*) against the scattering in the free atmosphere represented in Fig. 4 *b*. There is satisfactory agreement within the values $R > 10$.

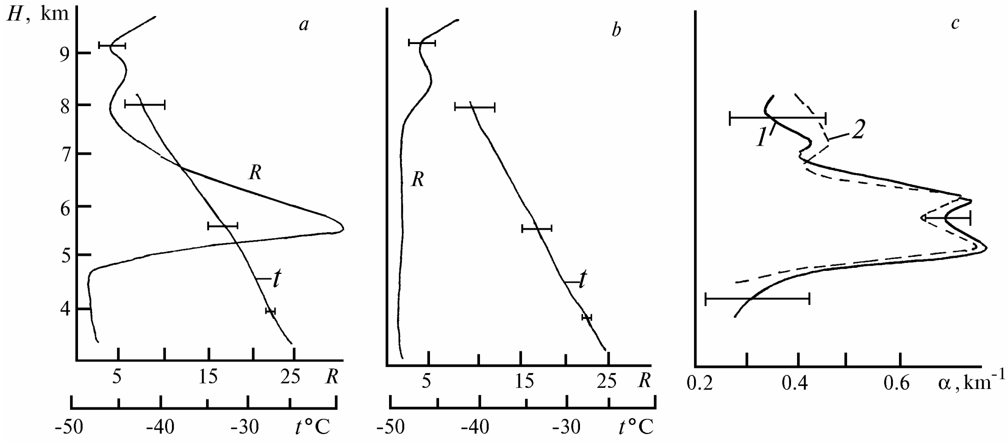

FIG. 4. *Profiles of scattering ratio R and temperature t corresponding to time:* 20 hr 15 min *(a) and* 21 hr 30 min *(b) of the temporal measurement of aerosol vertical distribution represented in Fig. 3; the extinction coefficient profile calculated by the formula (20) (1) and by comparing the profiles (a) and (b) under some assumptions (see the text) (2) (c).*

CONCLUSION

The described method of determining the coefficients of light backscattering and extinction shows significant promise for studying the troposphere and the stratosphere to the altitudes of 20 to 25 km.

The estimates indicate that the increase in the mean emitted energy to 10 W will make it possible not to exceed the relative error to 7 and 25% at the altitudes 5 and 15 km with spatial resolution 0.25 and 0.5 km, respectively. For the altitude interval between 15 and 25 km good estimates with spatial resolution of 2 to 3 km can be obtained.

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