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Vertical Ozone Profile Determination from Ground-based Measurements of Atmospheric Millimeter-Wave Radiation

K. P. Gaikovich*, E. P. Kropotkina, and S. V. Solomonov****
**Radiophysical Research Institute, ul. Lyadova 25/14, Nizhni Novgorod, 603600 Russia*
***Lebedev Institute of Physics, Russian Academy of Sciences, Leninskii pr. 53, Moscow, 117924 Russia*

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Abstract—An efficient technique for solving an inverse problem of remote sensing of atmospheric ozone at millimeter radio waves is described. The technique is based on an iteration scheme in which Tikhonov's method in the form of the principle of generalized residual is used at each step. A significant advantage of the method lies in the association of the regularization parameter and in the degree of smoothness of the solution with the total effective error. To increase the accuracy of the inverse-problem solution, a difference version of the technique is used. For this version of the technique based on data of thermal radio radiation of ozone molecules, the relative spectral behavior of brightness temperatures, but not their absolute values, are significant. Results of a numerical experiment modeling of ground-based atmospheric ozone observations within the spectral band centered at 142.2 GHz are given. Actual parameters, such as noise, operating frequencies, and spectra! resolution of the ozonometric instruments at the Lebedev Institute of Physics arc considered in computations. Results of the application of this technique to ozone observational data obtained in early 1996 over the Moscow region are given. The proposed method extends a class of problems, related to ecology and atmospheric physics, solvable with millimeter-wave-radiophysics methods.

INTRODUCTION

Investigations of the vertical ozone profile (VOP) in upper atmospheric layers not easily accessible is an important problem of ground-based remote sensing of the Earth's atmosphere at millimeter radio waves [1]. In solving this problem, the thermal radiation emitted by atmospheric ozone molecules at a rotational spectral line located within an atmospheric transmission band is recorded. Variously leveled layers, which are characterized by different atmospheric pressures and different ozone concentrations, contribute to this radiation. Within the millimeter range, line broadening is determined by collisions; i.e., it is proportional to the pressure. Therefore, the end shape of a spectral line contains information on the vertical ozone profile. To record this line, a receiving instrument of high sensitivity and high spectral resolution is necessary. Such an ozonometer has been designed at the Lebedev Institute of Physics, Russian Academy of Sciences, (FIAN) [1, 2]. A radiation sensor and a multichannel filter spectra! analyzer are components of this ozonometer. For solving the inverse problem lying in VOP retrieval from a measured spectrum, different theoretical models have been used in recent years [3-9]. It is well known that this inverse problem relates to ill-posed problems, because, in the absence of a priori information, a small experimental error results in an infinitely large retrieval error. A completely continuous operator corresponds to the direct problem, which lies in calculating the integral appearing in the equation of radiative transfer. An operator that is inverse to a completely continuous operator is known to be unbounded. Therefore, using available a priori information about the exact solution is necessary to solve the inverse problem. A preferable solution algorithm is determined by the information being used. This information may be statistical in character [8]. It may make it possible to use the optimum altitude-frequency discretization to linearize the integral equation and to specify its kernel [4]. The membership of the exact solution in any compact class of functions can also be used as a priori information [6]. In the last case, the solution can be obtained through minimization of the residual functional, since this functional is convex for compact functions. Notice that, in such a solution, points of discontinuity of the first kind are possible; it is also known that a function describing ozone distribution relates to the class of continuous functions that are not compact. Experience on solving problems with compact classes of functions shows that convergence of an approximate solution to the exact solution is very slow. This conclusion is confirmed by numerical modeling and VOP retrieval from the measurements of the spectral dependence of the optical thickness for the ozone line with resonance at 142 GHz [6].

The problem under consideration has a significant peculiarity that hampers an application of the majority of standard mathematical methods. This peculiarity lies in nonlinearity, i.e., in the dependence of the equation kernel on the ozone concentration profile. Our mode of solving is based on the iteration scheme of [10], in which Tikhonov's method in the form of the principle of generalized residual [11] is used at each step. This method uses superficial information on the smoothness of the exact solution.

This approach has a number of significant peculiarities that are shown to make our technique very efficient for solution of the above inverse problem of millimeter-wave remote sensing of atmospheric ozone.

STATEMENT OF THE INVERSE PROBLEM

Upon subtraction of the other atmospheric components, the relation between the brightness temperature T_B of thermal radio radiation and the VOP $U(h)$ can be represented as the integral

$$T_B(\nu) = \int_0^{\infty} U(h)K(U, \nu, \theta, h)dh, \qquad (1)$$

where $K = K(U, \nu, \theta, h)$ is the nonlinear kernel of the equation; ν is the frequency of radiation; and θ is the observation angle. To solve (1), we used the iteration scheme developed in [10]. According to this scheme, at each step of the iteration process, a solution is searched with Tikhonov's method of generalized residual [11]. At the first step, a model ozone profile $U^1(h)$ is substituted into the kernel of (1); at the second step, the retrieved profile obtained at the first step is substituted into the kernel, and so on. At each iteration, an integral linear Fredholm equation of the first kind is solved. With operators, equation (1) has the form

$$\mathbf{K}U = T_B^{\delta}, \qquad (2)$$

where T_B^{δ} is the data vector that is known with errors

$$\delta T_B^2 = \sup\|\mathbf{K}U - T_B^{\delta}\|_{L_2}^2 = \frac{1}{\Delta\nu}\int_{\Delta\nu}[T_B(\nu) - T_B^{\delta}(\nu)]^2 d\nu \qquad (3)$$

In (3), T_B is the right-hand side of (1), i.e., the exact solution $U(h)$, and $d\nu$ is the width of the frequency band under consideration. Within the context of this method, the error in the kernel can be taken into account. It includes a discretization error, inaccuracy in describing the absorption coefficient, and the errors due to disregarding temperature and pressure variations and to replacing the actual ozone profile in the kernel by a model profile. The measure of error in the kernel of (1) is specified as

$$\delta_h^2 = \sup\|\mathbf{K}_hU - \mathbf{K}U\|_{L_2}^2 = \sup\|\mathbf{K}_hU - T_B\|_{L_2}^2, \qquad (4)$$

where K_h is the operator corresponding to a preset approximate kernel in (2). The above errors can result in the incompatibility of the data vector with the equation being solved, because a smoothing action of the kernel of (1) limits the class of possible functions $T_B(\nu)$, so that the function T_B^{δ} can fall outside an admissible class in the presence of a random error. Of course, the measure of incompatibility δ_μ cannot exceed the kernel and measurement errors added together; i.e.,

$$\delta_\mu = \inf\|\mathbf{K}_hU - T_B^{\delta}\|_{L_2} \le \delta T_B + \delta_h. \qquad (5)$$

According to Tikhonov's method [11], an approximate solution U^{α} minimizes the smoothing functional

$$\mathbf{M}^{\alpha}(U) = \|\mathbf{K}_hU - T_B^{\delta}\|_{L_2}^2 + \alpha\|U\|_{W_2^1}^2. \qquad (6)$$

The notation $\|x\|$ means the norm of the x function as an element of the function space L_2 of square-integrable functions or the space W_2^1, the space of square-integrable functions having square-integrable generalized derivatives.

In particular, for the smoothing functional (6), we have

$$\|U\|_{W_2^1}^2 = \frac{1}{\Delta h}\int_{h_{\min}}^{h_{\max}}\left\{U^2(h) + \left[\Delta h\frac{dU}{dh}\right]^2\right\}dh,$$

where $\Delta h = h_{\max} - h_{\min}$ is the altitude interval on which the solution is sought.

The regularization parameter α to smooth an approximate solution is determined in Tikhonov's method [11] as a root of a one-dimensional nonlinear equation of generalized residual, which takes into account all the above components of the total error:

$$\rho(\alpha) = \|K_hU^{\alpha} - T_B^{\delta}\|_{L_2}^2 - \delta^2 = 0, \qquad (7)$$

where U^{α} is the function (solution) that minimizes functional (6), and $\delta^2 = (\delta T_B + \delta_h)^2 + \delta_\mu$ is the effective error parameter. This parameter takes into consideration the level of measurement errors, the inaccuracy in describing the kernel, and also the measure of incompatibility of equation (2) with its right-hand side, which depends on these uncertainties.

Thus, the regularization parameter a and, consequently, the degree of smoothness of the solution are related to the total effective error. This peculiarity is a significant advantage of the method. A very important advantage of the method of generalized residual over other methods lies in that, as the total error tends to zero in the integral metric L_2, the approximate solution converges to the exact solution in the metric W_2^1. This means that, according to the Sobolev enclosure theorem, this approximate solution converges uniformly, i.e., in the metric C, where the maximum modulus is the norm. On the other hand, unlike well-posed problems, the rate of convergence here is not proportional to a decrease in δ, but is slower. If the metric L_2 (square integrability) is used in the second term of (6), the convergence of the solution will also be in the metric L_2.

The parameters δ_h, and δ_μ can he determined by using numerical modeling in the course of minimizing (6). As a rule, the measure of incompatibility and the degree of kernel nonlinearity limit the level of residual by which functional (6) can be minimized.

Once corresponding discretization has been performed, the problem of minimization of a smoothing functional goes to its finite-dimensional analogue representing the quadratic programming algorithm well studied as a computational problem (see [11]).

The operator form of the iteration scheme [10] for the nonlinear problem described by (1) is as follows:

$$K^i U^{i+1} = T_B^\delta \qquad (8)$$

where the operator $K^i = K(U^i, \nu, h)$ and the model ozone profile is the null approximation $U^1(h)$, which can be described by an arbitrary function from W_2^1. It is known that the iteration procedure converges to the exact solution if the conditions of the principle of contracting mappings are fulfilled; i.e., a completely continuous operator (1) maps a closed convex set of Banach space onto itself [12]. The problem under consideration was solved numerically. Therefore, the convergence conditions were revealed with a numerical experiment like [5], where the iteration method was first designed for ozone sensing through limb measurements with satellites.

For application of the algorithm designed in [10], atmospheric ozone emission spectra given in absolute brightness temperatures are necessary. To obtain them, the measured radiation should be compared to the radiation of two blackbody thermal radiators of different well-known temperatures. It is difficult to introduce the exact correction for the difference between the radiation of regular and blackbody radiators. Therefore, a systematic error of such calibration is an inevitable source of additional errors in VOP retrieval.

To increase the accuracy of VOP retrieval, we used a differential version of the above method. For this version, data on the relative spectral behavior of temperatures rather than their absolute values are needed. According to this version, a VOP can be retrieved through differences between brightness temperatures measured in each of the channels of a radiometer, i.e., at each of the channel frequencies within the observational spectral line, and a brightness temperature measured in a reference channel. The reference channel was chosen so that its frequency was at the boundary of this spectral line. Such a procedure allows us to eliminate any influence of a systematic error of the absolute calibration. With this approach, equation (1) transforms to the equation

$$\Delta T_B(\nu) = \int_0^\infty U(h)\Delta K(U,\nu,\theta,h)dh, \qquad (9)$$

$$\Delta T_B(\nu) = T_B(\nu) - T_B(\nu_0),$$

$$\Delta K(U, \nu, h) = K(U, \nu, \Theta, h) - K(U, \nu_0, \Theta, h),$$

which is mathematically equivalent to (1). In (9),

$$\Delta T_B(\nu) = T_B(\nu) - T_B(\nu_0),$$

and $\Delta K(U,\nu,h) = K(U,\nu,\theta,h) - K(U,\nu,\theta,h)$,

where ν_0 is a fixed frequency.

A peculiarity of incorrect problems, such as a Fredholm equation of the first kind, lies in the lack of a definite relation between the error of the right-hand side of the equation and the accuracy of retrieval, since the error depends significantly on the form of the function under retrieval. Therefore, a study of characteristics of retrieval of typical model profiles such as the accuracy in relation to measurement errors, the number and frequencies of the channels, and so on can be performed through numerical closed-loop experiments and by no other way. Also, a numerical experiment is necessary to study convergence properties of the iteration procedure (8).

In applying the method under consideration, from the start, it is necessary to clarify the relation between the effective error parameter δ given by (7) and the predetermined characteristics of measurement errors, taking into account that the latter are random in their character. Let the error have a normal distribution with the mean $\Delta T_B(\nu)$ and standard deviation $\sigma_{TB}(\nu)$.

The error δT_B is considered to be the mean value of the integral in (3) rather than its maximum value. Then, we have

$$\delta T_B^2 = \frac{1}{\Delta\nu}\int_{\Delta\nu} \langle [T_B(\nu) - T_B^\delta(\nu)]^2 \rangle d\nu \qquad (10)$$

$$= \frac{1}{\Delta\nu}\int_{\Delta\nu} [\sigma_{T_B}^2(\nu) + (\Delta T_B(\nu))^2] d\nu.$$

It is seen that the determined error is split into the random and systematic terms. In the case under consideration, the first term of (10) is the radiometer sensitivity to fluctuations and the second one includes errors of the absolute calibration. It was shown above that in the differential version of the method, errors of the absolute calibration do not influence the result provided that they are equal throughout the frequency range being analyzed.

RESULTS OF THE NUMERICAL EXPERIMENT

The aim of the numerical experiment lay in determination of the accuracy of preset model VOP retrieval, and of the degree of influence of uncounted errors in the emission spectra of atmospheric ozone and in the vertical temperature and pressure profiles on the accuracy.

We also studied retrieval error determined by a numerical implementation of the method; this error is hereafter called the error of the method. Below, results of numerical experiments modeling ground-based observations of the ozone layer at a zenith angle of 60° and at frequencies of a spectral line centered at 142.2 GHz are presented. It is known that this well-marked ozone spectral line lying within an atmospheric transmission band is easily registered by ground-based radiometers [1,2]. With methods of radar astronomy, inevitable tropospheric attenuation caused mainly by absorption of radiation by oxygen and water vapors are measured and taken into consideration [2]. In the following computation, we take that the spectral line under consideration is corrected for the tropospheric attenuation and thus it is converted to the conditions of observation from outside the troposphere. In the numerical experiment, the atmospheric ozone emission spectrum was computed using parameters of the model of stratified atmosphere ranging from the Earth's surface to a 100-km altitude. The vertical profiles of temperature, pressure, and ozone content were set, the ozone content being given as the mixing ratio, ppm. The flexibility of the model makes it possible to vary the parameters. In computations, such parameters of the FIAN instrument [1, 2, 13] as the noise, the operating frequencies, the quantity of frequency channels, the range of frequencies in the analyzed spectral band, and the spectral resolution were taken into account.

The procedure of the numerical experiment included computations of the atmospheric ozone emission spectrum from the equation of radiative transfer with the above parameters of the model multi-layer atmosphere. To study the influence of the instrumental noise, the value of smearing of a computed spectrum by this noise was set as a function of the integration time of the useful signal. The inverse problem to be solved consisted in VOP retrieval from the spectrum shape. To determine the retrieval error, the VOP obtained as a result of solving the inverse problem was compared with the corresponding preset model VOP.

The error component δ_h caused by kernel uncertainty due to discretization was estimated through the comparison of brightness temperatures calculated with two procedures, namely, from the model and from the exact integration using simple profiles. The estimated error was well below the radiometer sensitivity δT_B. In the algorithm, the error component caused by discretization, $\delta_h = 0.001$ K. To obtain this result, we used almost the entire memory capacity and the internal performance of our personal computer. The above value of the error can be considered as the accuracy limit of the numerical scheme that can be implemented with our technique.

The numerical experiment on estimation of the measure of incompatibility δ_μ at the minimization of (6) showed that, to a first approximation, nonlinearity of the kernel does not lead to an incompatibility with the vector of brightness temperatures when the actual VOP is replaced by the model VOP. Thus, from (5) it follows that in the algorithm under consideration, $\delta_\mu < \delta T_B$. Taking into account that the error estimated from the maximum of (3) should be factually higher than the mean error (10), it is reasonable to set:

$$\delta_\mu = \delta T_B. \tag{11}$$

As was noted above, the principle of generalized residual expressed in (7) contains a definite arbitrariness. This fact fully justifies the description of the error by (10) and the introduction of condition (11). In numerical modeling, when a VOP should be retrieved from the data vector computed with the same VOP, the condition $\delta_\mu = 0$ is fulfilled. Let observational data be computed or random errors be added to computed T_B values. Thus, according to (11), the total error δ, with possible incompatibility taken into consideration, must be $\sqrt{2}$ times higher than the standard deviation corresponding to the fluctuation sensitivity if the condition $\delta T_B >> \delta_h$ is fulfilled; i.e.

$$\delta = \sqrt{2}\delta T_B = \sqrt{2}\sigma_{T_B}. \tag{12}$$

If a lower error is used in solving the problem, there is reason to obtain VOP peculiarities that do not exist in reality. In contrast, if enhanced errors are used in computations, actual features of VOP can be smoothed. Thus, the parameter 8 (12) determined by the above components of measurement errors is optimum.

The widely used approach to estimation of the accuracy of the method lies in determination of the rms retrieval error for a representative statistical ensemble. In this work, the above property of uniform convergence of the method can be used. Owing to uniform convergence of the method, we can use the maximum modulus of the difference between numerically retrieved and preset VOPs as a measure of accuracy of a solution. The quality of a solution depends on the degree of complexity of a preset VOP. Therefore, it is expedient to obtain estimates of retrieval accuracy for both typical and extremum profiles. To estimate the limiting possibilities of the method, it is desirable to study the accuracy of VOP retrieval not only at a practically achieved level of measurement errors, but also at the lowest error, $\delta = \delta_h = 0.001$ K, possible at the numerical implementation of the method.

As the preset profiles in examples given below, we used three VOPs: (1) is characterized by a mesospheric maximum and an enhanced stratospheric ozone content, up to 11.7 ppm at the 35-km altitude; (2) is characterized by a lowered ozone content, down to 3.7 ppm at the 35-km altitude; (3) is characterized by a moderate ozone content, 8.7 ppm at the 35-km altitude. As the null approximation in the iteration scheme, we used a VOP characteristic for the midlatitude Northern Hemisphere (Fig. la). The quality of retrieval may be char-

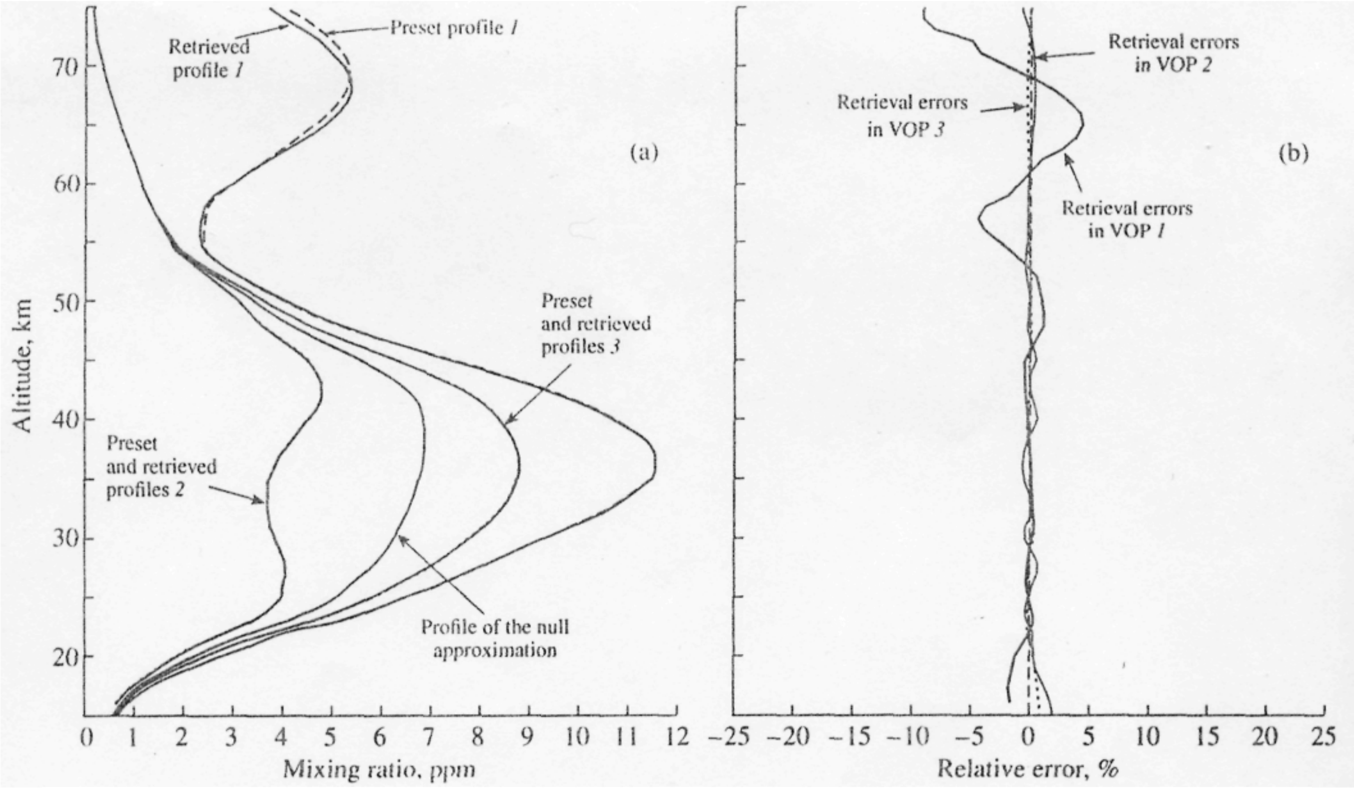

Fig. 1. (a) Retrieval of three preset VOPs. For VOPs 2 and 3. the plots of retrieved and preset profiles practically coincide. (b) Retrieval errors in the VOPs of Fig. 1 a.

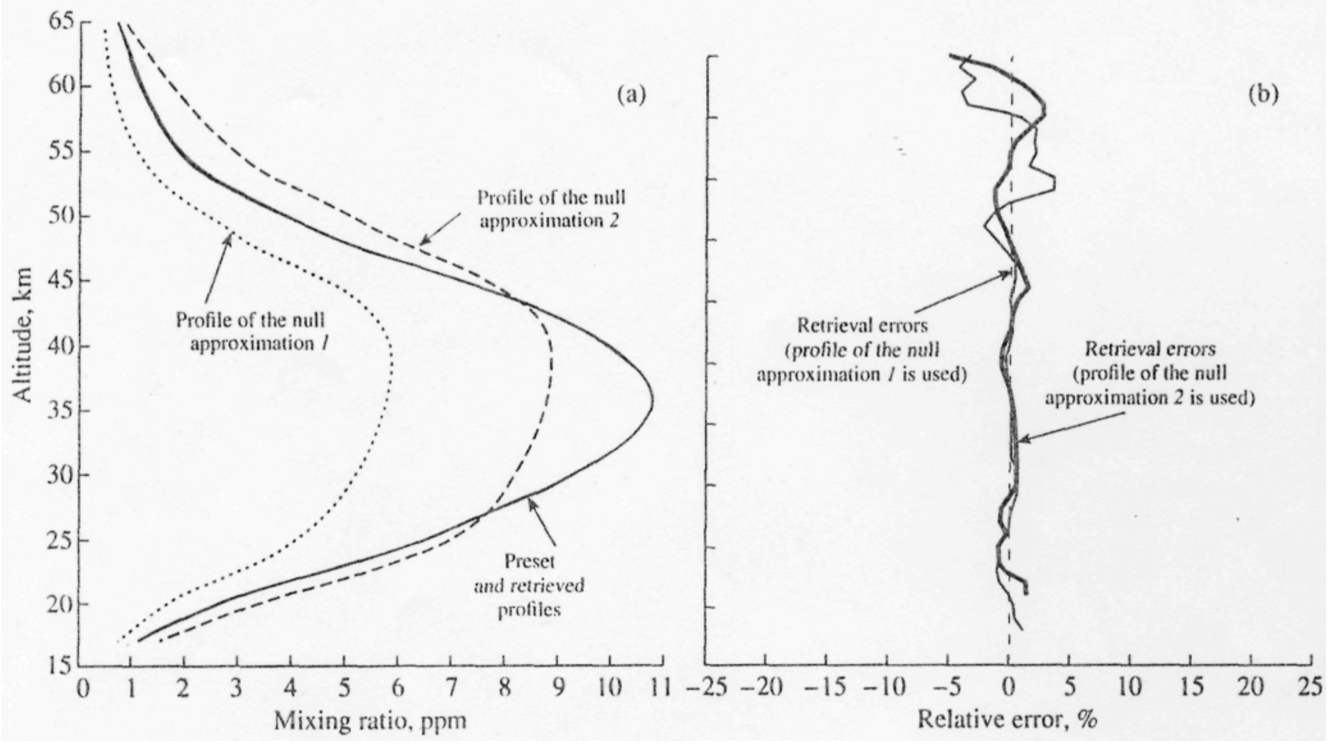

Fig. 2. (a) VOP retrieval for two preset profiles used ns the null approximation. The plots of retrieved and preset profiles practically coincide, (b) Retrieval errors in the VOP of Fig. 2a for two profiles of the null approximation.

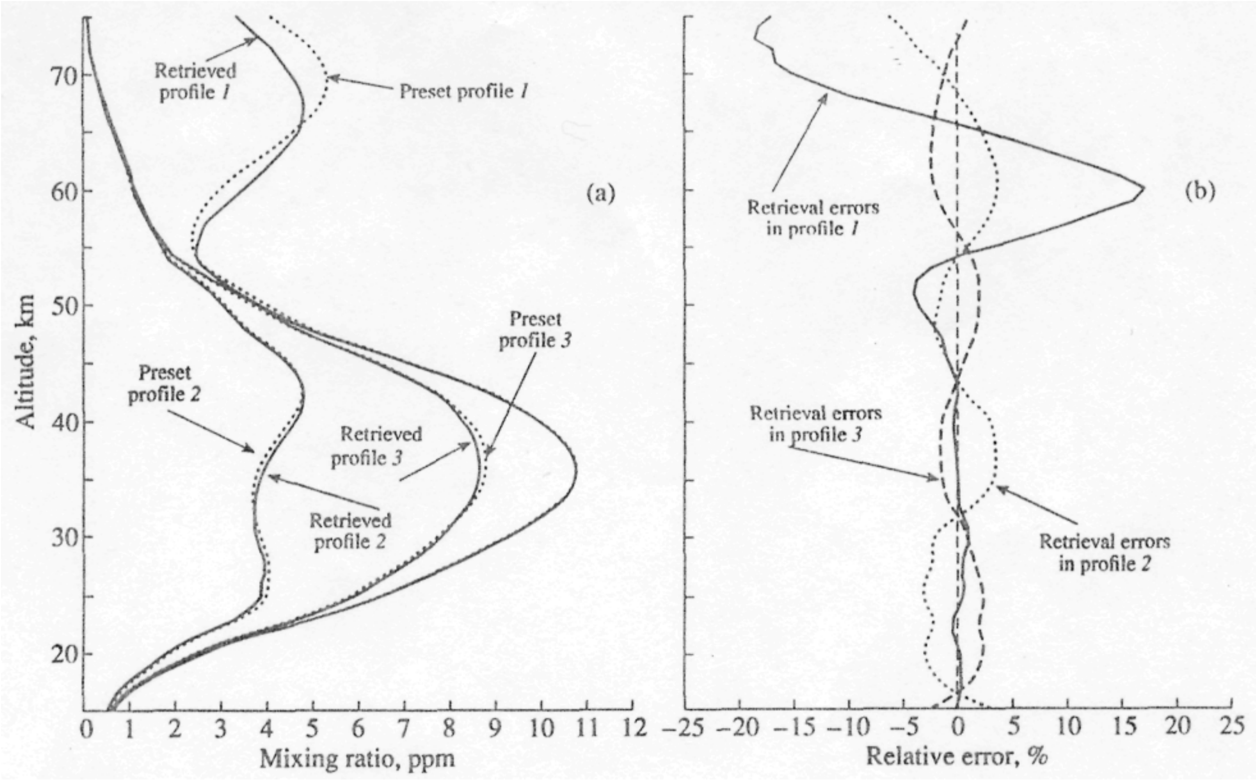

Fig. 3. Influence of the radiometer noise on the errors of VOP retrieval. Double-band noise temperature, 400 K; integration time, 1 h: (a) Preset and retrieved VOPs, (b) Retrieval errors.

acterized by the relative difference between a retrieved profile and a preset profile:

$$\Delta = \frac{U^{\alpha} - \tilde{U}}{U} \times 100\%.$$

To illustrate the limiting possibilities of the method, at $\delta = \delta_h = 0.001$ K, Fig.la gives results of VOP retrieval for three above preset VOPs. In this figure, the retrieved profiles are given as solid curves. They virtually coincide with the preset VOPs given as dashed curves. Figure Ib gives the errors of VOP retrieval. The results of our computations showed that the limiting errors of the method do not exceed 1-2% at altitudes from 20 to 50 km and 10% at altitudes from 15 to 20 km and from 50 to 75 km. Numerical experiments showed that the form of the null approximation influences the retrieval accuracy only slightly. It is seen from the examples given in Fig. 2 and relating to two preset VOPs used as the null approximation.

The influence of the instrumental noise on the results of VOP retrieval is seen from Fig. 3. The noise tailing of the ozone spectral line calculated from a preset VOP was specified in a program on the basis of the double-band noise temperature achieved in the FIAN radiometer and equal to 400 K. In computations, the number of spectral channels, 80, and the frequency bandwidth of the radiometer, 260 MHz, were specified. Figure 3a gives the retrieved VOPs. They correspond to 5 = 0.04 K implemented in the FIAN radiometer at a signal integration time of 1 h. The retrieval errors are presented in Fig. 3b. The computations show that the total error of VOP retrieval caused by the errors of the method and by the noise of the cooled instruments does not exceed 2-3% at an integration time of 1 h. This result is valid in an altitude range at least from 20 to 50 km. This error can be reduced through prolongation of the integration time and enhancement of the sensitivity of instruments. The computed error is significantly lower than the errors in other methods [4,7-9] used in leading foreign observatories for ozone observations at millimeter waves.

An analysis of the influence of errors in preset temperature and pressure profiles on the errors of VOP retrieval was performed. For this purpose, we carried out computations not only with model vertical temperature and pressure profiles but also with profiles of the same parameters observed under extreme conditions, for example, for a deep-dropping stratospheric polar vortex whose influence may extend to the mid-latitude stratosphere, including the stratosphere over Moscow. Such profiles sometimes significantly differ from the climatic average. Aerological sounding over the Moscow region shows that sometimes a 10-mbar-pressure level drops by 1 km below the average, and the temperature at altitudes from 20 to 50 km decreases by 20°C below the average. The numerical experiment showed that the use of average temperature and pressure pro-

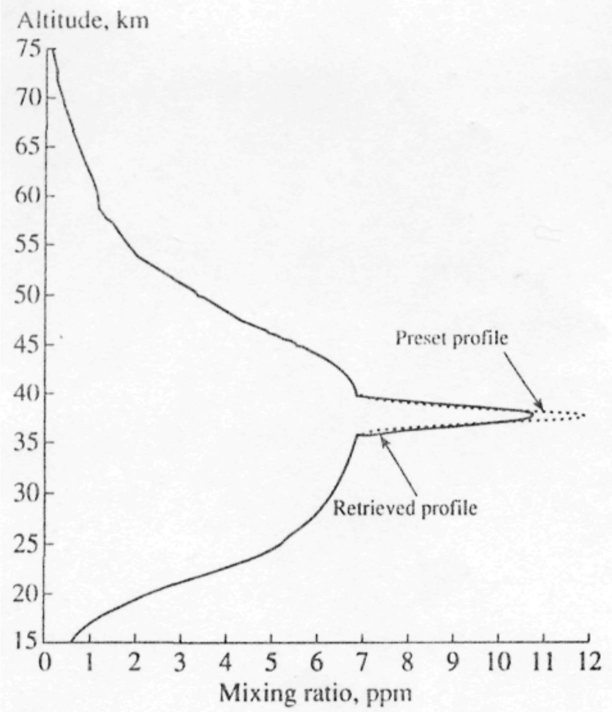

Fig. 4. Retrieval of VOP with a peculiarity at an altitude of about 40 km.

files in the inverse problem of VOP retrieval leads to errors up to 10-15%.

During cold periods, for example, at a deep-dropping stratospheric polar vortex, atmospheric parameters significantly differed from the average ones. According to the aforesaid, in VOP retrieval from spectra obtained during cold-period ozone observations, the temperature and pressure profiles should be corrected with the aerological sounding taken into account, and it is reasonable to extrapolate the radiosonde data to higher atmospheric layers. The extrapolation is justified by winter-season intensification of temperature connections between atmospheric layers [14]. As estimates show, such corrections can decrease the error of VOP retrieval down to 5-6% at a signal integration time of 1 h.

The data on the resolution capability of the method are shown in Fig. 4, where the results of retrieval of a peculiar VOP are given. The peculiarity of the VOP lies in a thin altitude one-layer structure described by the Gaussian distribution $\Delta U = A\exp[-(\frac{h-h_{\max}}{\Delta h})^2]$ where A = 5 ppm, $h_{\max}$ = 38 km, and Δh = 1 km. The computations were performed at the above minimum error. The computational procedure was broken into two stages. At the first stage, we solved the inverse problem within altitudes from 15 to 75 km. At the second stage, the altitude interval was limited by the region of localization of one-layer structure, this region being determined at the first stage. Such a version allowed us to retrieve more exactly the inhomogeneous structure. Then, the results obtained at the two stages were sewn together. The resulting retrieved VOP profile is shown in Fig. 4.

SOME RESULTS OF MILLIMETER-WAVE MEASUREMENTS

The above technique was used in the processing of ozone observational data obtained in Moscow with the FIAN radiometer. VOPs were retrieved from atmospheric ozone emission spectra measured at 142.2 GHz. In solving the inverse problem with this technique, we used vertical temperature and pressure profiles that were kindly put at our disposal by the Russian Hydrometeorological Center. The retrieving program involves a computation of the spectral line from a retrieved VOP and a comparison of this line with the measured line. In the examples given below, distinctions between computed and measured spectral lines do not exceed a measurement error caused by instrumental noise.

As examples, Fig. 5a presents ozone spectral lines recorded on January 26 and February 9, 1996.

The corresponding retrieved VOPs are depicted in Fig.5b.

The data measured with the same radiometer in January-April 1996 and then processed with the above technique allowed us to obtain new results and to reveal important peculiarities in VOPs. We found that, over January and over some periods of February and March, the ozone content in the middle and upper atmosphere was stable and held 2 times lower than the mode! ozone content [15]. The decreased ozone content is evidently caused by a stable stratospheric polar vortex that influenced the stratosphere over the Moscow region. According to the data of the Central Aerological Observatory, during the period under consideration, the total ozone content over Moscow was about 15% below the average.

A similar decrease in the atmospheric ozone content along the vertical direction had also been observed before, for example, in January 1989. However, earlier it was not pronounced as strongly [2].

Another peculiarity of the winter season of 1996 lies in an increase of the ozone content in the beginning of February. Figure 5b gives an example of a VOP retrieved from the observational data of February 9, 1996. This increase in ozone content, which was also recorded in optical measurements of the total ozone content over Moscow, was due to the Mediterranean stratospheric anticyclone, which delivered ozone-enriched air into this region.

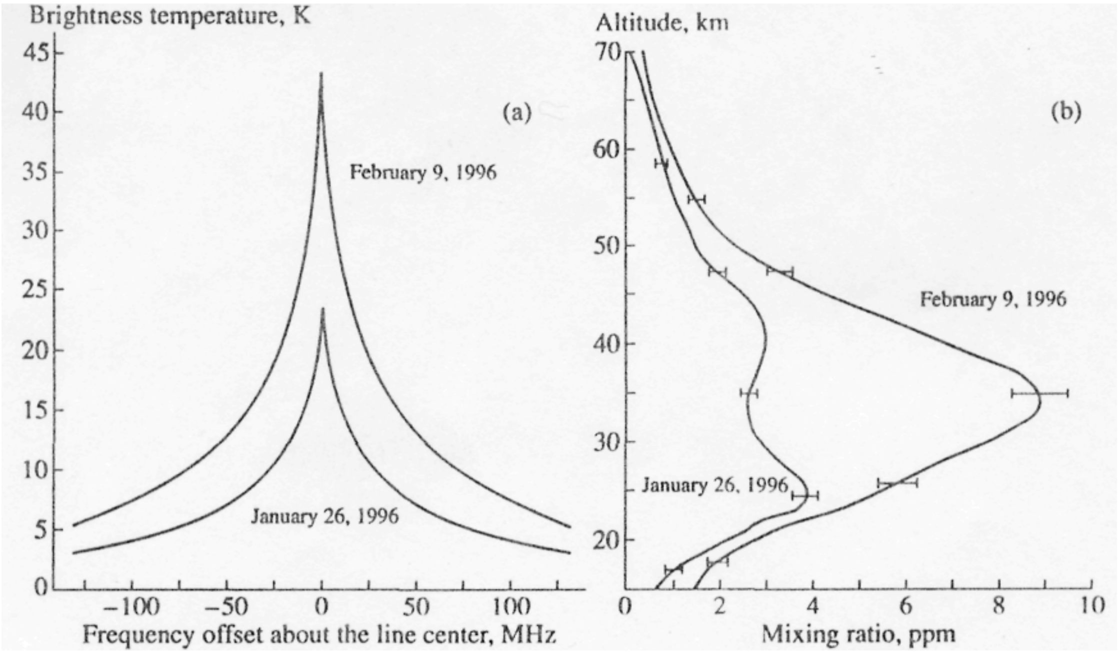

Fig. 5. (a) Atmospheric ozone emission lines centered at 142.175 GHz; January 26 and February 9, 1996; elevation angle, 30^{0}; integration time of about 1 h. (b) VOPs corresponding to the spectra of Fig. 5a.

CONCLUSION

Our investigations show that millimeter-wave remote sensing of the atmospheric ozone layer is highly efficient. The application of the method of generalized residual to processing recorded ozone spectra improves the accuracy of VOP retrieval and extends the range of altitudes within reach of sounding. Employing this method extends the class of problems of ecology and atmospheric physics that are solvable with radio-science methods based on the use of millimeter waves.

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