MOLECULAR WATER VAPOR ABSORPTION OF RADIATION IN THE 8–13 μm ATMOSPHERIC RELATIVE TRANSPARENCY WINDOW

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Generalized results of long complex laboratory and field studies, extending over many years, on the water vapor continuum and selective absorption of CO_2-laser radiation for a wide range of variation of the atmospheric parameters are given. A refined model of the water vapor continuum is developed which takes account of all of the continuum regularities and is in good agreement with the laboratory data available elsewhere and our own field observations as well.

INTRODUCTION

Absorption in the 8–13 μm atmospheric relative transparency window is the main factor determining the magnitude of the thermal emission from the Earth into space, i.e., it plays an important role in shaping the heat budget of our planet. The first studies of the 8–13 μm-window phenomenon date back to the end of the last century, and it was at the beginning of the present century that absorption in the window was associated with water vapor.[1] In 1938 Elsasser[2] put forth the hypothesis of the formation of this absorption in the form of a continuum caused by the far wings of an oscillating rotatory ν_2 and purely rotatory bands of the water vapor absorption with centers at about 6.3 μm and beyond 14 μm, respectively. According to Elsasser[2] the water vapor optical depth is given by the product of the total moisture content and the absorption coefficient, which linearly depends on the total pressure and weakly increases with temperature. Further studies under field conditions led at the beginning of the sixties to the paradoxical conclusion of the possibility of the occurrence of negative optical thicknesses at low humidities.[3] To remove this paradox the idea was proposed in Ref. 3 of the nonlinear dependence of the continuum absorption on the humidity. A qualitatively new stage in the investigation of the 8–13 μm window began at the end of the sixties and continuing into the first half of the seventies. It was facilitated by the appearance of the opportunity of using CO_2 lasers as radiation sources both in laboratory experiments in long-baseline multipass cuvettes and in long paths under field conditions, whose generation spectrum on various isotopes of carbon dioxide in the bands 00°1–10°0 and 00°1-02°0 over laps a large part of the 8–13 μm window. Moreover, broad perspectives of the use of these lasers in solving various scientific and technical problems, e.g., remote atmosphere sensing, optical communication lines, optical location, and so on, have required more reliable information about molecular absorption in the 8–13 μm range. Thus, selective absorption, insignificant in comparison with continuum absorption for the radiation from the Sun, the Earth, and classical heat sources, proves to be rather important for laser radiation if an overlap of the generation and absorption lines takes place, even if it is only partial.

Under actual atmospheric conditions the investigation of the processes of water-vapor molecular absorption is complicated, first, by the relative smallness of the absorption, second, by the simultaneous action of a number of factors which are not subject to precise separation and control (temperature, pressure, absorber concentration, nonuniformity of the medium, etc.), and, third, by the superposition of aerosol extinction and molecular absorption of other gaseous components of the atmosphere. It is for this reason that during the last 20 years much attention has been focussed on laboratory experiments involving optical multipass cells, in which it is possible to vary and strictly control the content, temperature, and pressure of the medium being analyzed as well as the concentration of the absorbing gas. The analysis already of the results of the first laboratory studies[4–6] has not only verified the nonlinear dependence of the continuum absorption on the water-vapor content but has also revealed a negative dependence of the absorption on the temperature, i.e., a decrease in the absorption with increase in the temperature.[5,6] Such a form of the temperature dependence allowed Bignell[6] and Varanasi et al.[7] to advance the dimer hypothesis of the mechanism of formation of the water vapor continuum in the 8–13 μm window.[6,7]

From the second half of the seventies besides the multipass cells used to perform laboratory experiments, laser optical-acoustic spectrophones have begun to be used.[8] The simplicity of construction and the ease of processing the measurement results have attracted the attention of many researchers, who have carried out measurements of the water vapor absorption coefficients for dozens of generation lines of

several carbon dioxide isotopes.[9–13] The spectrophones drawbacks, associated with the necessity of first calibrating their dependence on laser power and the temperature of the medium lead, however, to large systematic measurement errors.[13,14] Owing to this the published measurement data involving spectrophones frequently differ noticeably among themselves and from the measurement results obtained using multipass cells.[12,12,15–17] This circumstance together with a number of qualitative differences in the results of various groups motivated us to carry out complex laboratory and field investigations of molecular (selective and continuum) water vapor absorption of radiation from a tunable CO_2-laser, the results of which are presented in the present paper.

EQUIPMENT AND METHODS

Laboratory investigations of molecular water vapor absorption were carried out using an apparatus complex based on the optical multipass cell, connected in a White circuit.[18] The baselength of the multipass cell, equal to 50 m, enabled us to reach thicknesses of the studied gas layer of up to 3 km. In the multipass cell it was possible to regulate and control pressure variations from 10^{-7} to 5 atm and temperature variations from 284 to 353 K. Special devices maintained a medium of pure water vapor in the multipass cell or a mixture of it with nitrogen (dehumidified air).[19]

A universal, tunable CO_2 laser, assembled according to the conventional scheme, served as the radiation source. The cavity was formed by a spherical gilt mirror and a diffraction grating operating in the self-collimating mode with feedback in the first order and radiation removal in the zeroth order of the grating spectrum. Self-tuning of the radiation frequency is realized using a piezoelement. By changing the working mixture it was possible to obtain generation on the lines of three isotopes: $^{12}C^{16}O_2$, $^{13}C^{16}O_2$ and $^{12}C^{18}O_2$ (Ref. 20).

A liquid-nitrogen cooled, gold-alloyed germanium-based photoresistor served as the radiation detector,the electrical signal from which was recorded by an appropriate device hooked up to a microcomputer.

A completely identical experimental scheme was realized under field conditions. The source and radiation detector together with- the White mirrors were located indoors, and the path laid between them. The only difference consisted in placing the radiation detector in an integrating sphere to reduce the effect of air turbulence.[21]

The value of the water vapor transmission of the laser T_2 radiation was found from the usual ratio for a two-ray scheme of the intensities of the radiation passing through the multipass cell with the investigated gas and an evacuated one, normalized to the source power.

WATER VAPOR CONTINUUM

The results of the investigations of the continuum using multipass cells[4–6] were generalized in Ref. 15 and represented in the form of the following model:

$$\tau_\lambda = LC_s\omega[p+\gamma(P-p)]; \quad (1)$$

$$C_s = C_s(\theta=296)\ \exp[\theta_0(1/\theta-1/296)]; \quad (2)$$

$$C_s(296) = a+b\ \exp(-\beta\nu), \quad (3)$$

where $\tau_\lambda = -\ln T_\lambda$ is the optical thickness; L is the path length; p and P are the partial pressure of water vapor and the total pressure of the medium; ω is the amount of molecular water vapor per unit volume; θ is the medium temperature; $\theta_0 = 296$ K is the initial temperature at which the absorption coefficient C_s is measured; ν is the wave number; $\nu = 1/\lambda$, λ is the wavelength; and, the remaining parameters are adjustable parameters.

Approximations (1)—(3) are included in the widely used and continuously updated computer program package for calculating the absorption of radiation in the atmosphere, LOWTRAN,[22] to calculate the continuum water vapor absorption. The results of the subsequent laboratory and field tests are, as a rule, compared with the calculations using Eqs. (1)–(3), i.e., with the data of Refs. 4–6 or Ref. 9, which give the values of C_s and γ for 27 lines of the $^{12}C^{16}O_2$ laser, measured simultaneously with the help of a spectrophone and a multipass cell. An alternative continuum model was proposed in our papers of 10–15 years ago[23–26]:

$$\tau_\lambda = Lf(\lambda)[k_1a(1+\alpha P)+k_2a^2\cdot\exp(-\Delta H/R\theta)]; \quad (4)$$

$$f(\lambda) = b_0+b_1\cdot\exp(-b_3/\lambda), \quad (5)$$

where L is the path length; a is the absolute humidity; P is the total pressure; θ is the temperature; and, the rest are adjustable parameters.

A comparison of the data of different authors (Refs. 13, 15, 16, 17, 26) shows that for high humidities and long paths the values of transmission calculated according to the approximate expressions and according to the data of different authors differ by 20–30%. The basic difference between approximate expressions (1) and (4) lies in the forms of their dependence on the water vapor concentration and the total pressure. Moveover, due to the multimodality of the CO_2 laser, with which the experimental data for approximation (4) were obtained, formula (4) gives absorption values which are too high by 7–15%. (The laser spectrum included the P16 line, which is subject to noticeable selective water vapor absorption). These circumstances motivated us to carry out new experiments.

THE IMPROVED CONTINUUM MODEL

The choice of the specific laser lines at which to investigate the continuum was made on the basis of a preliminary analysis of the absorption spectrum in such a way that in the vicinity of the lasing frequencies of the CO_2 lasers there were no noticeable water vapor

absorption lines. Altogether 76 lines of three isotopes of carbon dioxide $^{12}C^{16}O_2$, $^{13}C^{16}O_2$ and $^{12}C^{18}O_2$ were chosen. In the experiments the absolute humidity varied from 0.5 to 25 g/m^3, the total pressure – from 0 to 1.6 atm, and the temperature – from 284 to 353 K. The path length was more often than not 3 km.

Before proceeding to examine the results of specific experiments,we will make one remark. In the field and laboratory experiments using multipass cells we measured the value of the transmission of radiation. The same transmission value is used in solving most problems of atmospheric optics and geophysics which involve an analysis of the propagation of radiation in the atmosphere. Even the measurement of the absorption coefficient by the optical-acoustic method is based on a calibration against a known absorption coefficient, which is found from the measured transmission of the appropriate radiation with the appropriate absorber. That is why in what follows the results and their analysis will, as a rule, be presented in terms of transmission values.

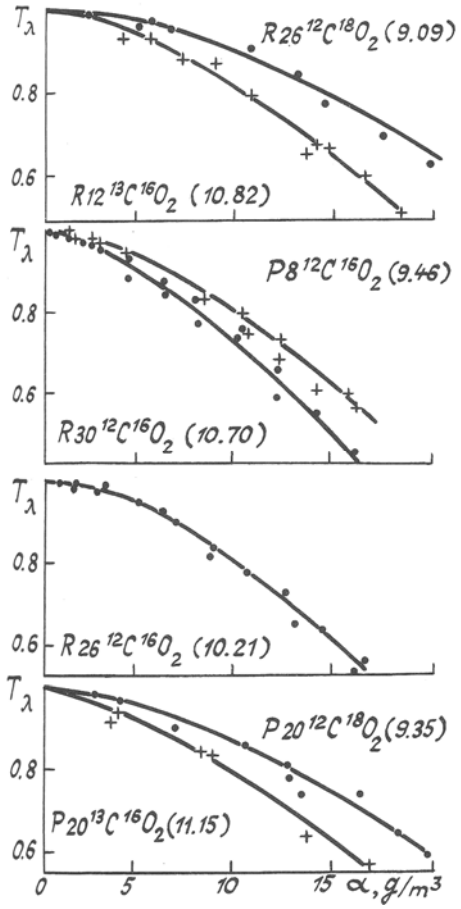

FIG. 1. Transmission of CO_2–laser radiation by water vapor. Radiation wavelength (μm) is given here and below in parentheses.

Results of measuring the transmission of radiation of different lines of a CO_2–isotope laser through pure water vapor over a 3 km path at constant temperature are given in Fig. 1. The analysis of the measurement' results shows that the dependence of the absorption of the CO_2 laser radiation by water vapor on the water vapor partial pressure has the same character over the entire investigated wavelength range from 9 to 11 μm. The absorption of the CO_2 laser radiation in this wavelength interval depends nonlinearly on the partial pressure.

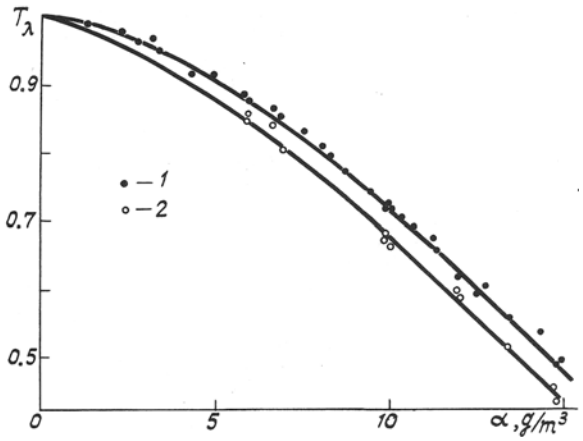

FIG. 2. Transmission of radiation at the line P20 (10.59 μm) of a CO_2–laser by water vapor and a mixture of it with nitrogen at a total pressure of 1 atm: 1) pure water vapor and 2) water vapor with nitrogen.

The obtained data were processed by the method of least squares, assuming various forms of the dependence of the absorption on the humidity. The best result was obtained for a dependence of the form (4) with $a > 1.2$ g/m^3 and $\theta = 293$ K and with $a > 2.6$ g/m^3 and $\theta = 323$ K the quadratic part being greater than the linear one, i.e., the difference from a purely quadratic dependence of the type (1) obtained in Refs. 4–6 is not great. The reason for the nondetection in Refs. 4–6 of the small linear part is evidently that the optical paths realized in them are 3–6 times shorter than those realized in our work. All researchers have noted that the broadening pressure of the nonabsorbing gas weakly affects the value of the water vapor continuum absorption and that the evaluation of quantitative values is associated with technical problems. It is for this reason that all of the previous experiments were carried out with a broadening pressure of 1 atm, which did not make it possible, as been mentioned above, to evaluate sufficiently unambiguously its quantitative value. The longest path realized in the present work made it possible to carry out a measurement at different pressures of the broadening gas and different temperatures. An example of the result obtained is shown in Fig. 2. Thus, at a constant temperature of 293 K and the humidity around 10 g/m^3 the transmission of radiation by a mixture of water vapor and nitrogen is decreased by 2.5–9% in comparison with pure water vapor with the broadening pressure

increased to as high a value as 1.3 atm. In other words, at a constant water vapor concentration increasing the broadening gas pressure by about 50 times changes the transmission by only a few per cent, which is many times less than what follows from the dispersion contour of the absorption lines.

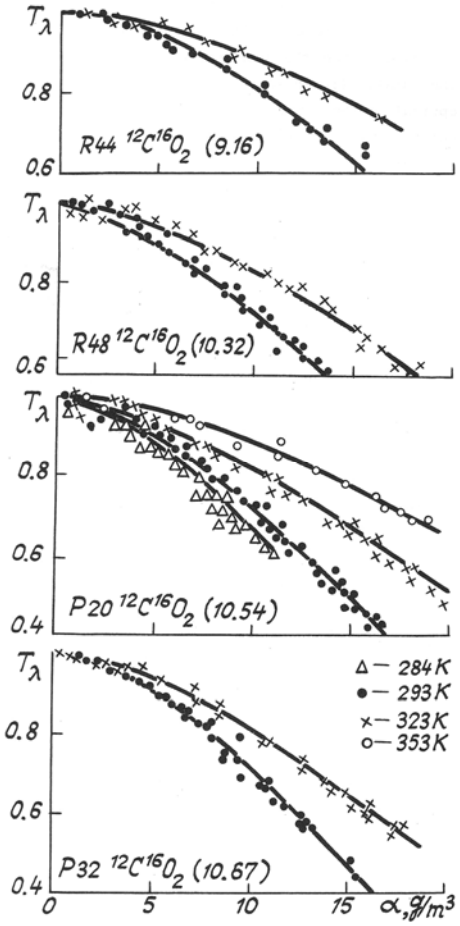

FIG. 3. Transmission of CO_2–laser radiation by water vapor at different temperatures.

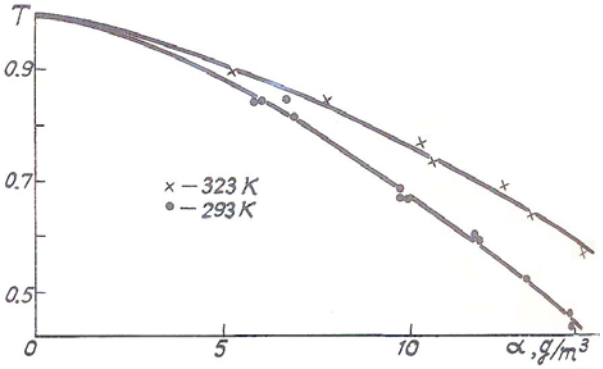

FIG. 4. Transmission of radiation at the line P20 (10.59 μm) of a CO_2–laser by water vapor with nitrogen at different temperatures.

The results of measuring the transmission of radiation at some CO_2–laser lines at different temperatures are given in Figs. 3 and 4. As can be seen from Figs. 3 and 4, the transmission increases as the temperature increases. In addition, a thorough analysis of numerous!, measurement data showed that in Eq. (4) not only K_2 but also K_1 depends on temperature. Moreover, unlike K_2, K_1 increases as the temperature increases. This experimental fact agrees with the predictions of Ref. 6 and Ref. 24 concerning the temperature dependence of K_1 and K_2, and may also explain deviations from Eq. (2) of the temperature dependence, observed at $\theta > 400$ K in Ref. 27.

The CO_2–lasers (operating on different lines of three carbon dioxide isotopes) used in the present work cover the 9–11 μm wavelength interval and make it possible to study the spectral dependence of the continuum. Taking into account the fact that the dependence of the absorption! continuum on the water vapor concentration, the pressure, and the temperature over the entire studied range remains the same, it is possible to represent τ_λ in the form $\tau_\lambda = f(\lambda)/\tau_{10.6}$ (τ_λ and $\tau_{10.6}$ are the optical thickness at the wavelengths λ and 10.6 μm, respectively; $f(\lambda)$ is a factor which depends on the wavelength and is equal to unity at $\lambda = 10.6$ μm). Guided by the predictions of theoretical studies of the exponential spectral dependence of the absorption,[28] we found that $f(\lambda)$ can be adequately represented by the following approximation:

$$f(\lambda) = b_0 \cdot \exp\left(\frac{\lambda^3}{b_1 + b_2\lambda^3} \right), \tag{6}$$

where $b_0 = 0.297$, $b_1 = 800$, and $b_2 = 0.15$. In terms of the above-described relationships the formula of the refined water vapor continuum model can be represented in the form:

$$T_\lambda = \exp\left\{ f(\lambda) L [k_1 a \theta^n (1 + \alpha P) + k_2 a^2 \exp(H/\theta)] \right\}, \tag{7}$$

where a is the absolute humidity in g/m^3, P is the total pressure in atm; θ is temperature in K, L is the path length in km; $f(\lambda)$ is given by formula (6); and, λ is the wavelength in μm.

The adjustable parameters have the following values:
$k_1 = 0.22 \cdot 10^{-6}$ m$^3 \cdot$ g$^{-1} \cdot$ degree$^{-3/2} \cdot$ km^{-1},
$k_2 = 0.82 \cdot 10^{-6}$ m$^6 \cdot$ g$^{-2} \cdot$ km^{-1}, $n = 1.5$,
$L = 1.95$ atm^{-1}, and $H = 2066$ K. Calculations based on Eq. (7) are shown in Figs. 1–4 by the solid lines.

This model, like the previous one (4), describes the exponential dependence of the continuum absorption on the wavelength, the nonlinear dependence on the humidity, and the weak dependence on the total pressure. But the temperature dependence is of a more complicated nature: the term in formula (7) that is quadratic in the humidity has a negative dependence on temperature; that is, the absorption decreases as the temperature increases, while the linear term has a positive dependence.

It is not possible on the basis of the proposed model of the water vapor continuous absorption to determine the mechanisms of formation of this absorption unambiguously. The experimental facts stated in the present paper are, to a certain extent, only an argument for the hypothesis of the action of two absorption mechanisms in the spectral range 8–13 μm. The first of these, described by the second part of expression (7), indicates that an important role is played by absorption by pairs of closely colliding absorbing molecules (in the extreme limiting case by dimers). This is indicated by the nonlinear dependence of the absorption on the water vapor pressure, the relatively strong negative temperature dependence of the continuous water vapor absorption, the closeness of the adjustable parameter H $\left(H = \frac{\Delta E}{R}\right)$ to the value of the bond energy of the water vapor dimer ΔE and the weak dependence of the absorption on the pressure of the broadening gas.

The second mechanism is characterized by a linear dependence on the humidity, a relatively weak positive temperature dependence, and a relatively strong dependence on the total pressure — the first part of the expression (7). Such a mechanism is characteristic of wide collisions of absorbing with nonabsorbing molecules, i.e., for the near wings of the weak water vapor absorption lines, values of which are available in the 8–13 μm region.

The model of the continuous water vapor absorption (7) is applicable, strictly speaking, only to the conditions of variation of the physical parameters, under which the laboratory experiments were carried out. In addition, the values of the water vapor content and the pressure of the broadening gas realized in these experiments exceed the conditions occurring in the Earth's troposphere. The experiments do not include the region of negative Celsius temperatures, and, in order to extend the given model to these temperatures, an additional justification is needed, which is also necessary for the extrapolation of the data obtained in the 9–11 μm spectral range, to the whole 8–13 μm atmospheric window. This can be done by comparing the model with the data of other laboratory experiments.

Before making this comparison, it is necessary to state some considerations as to ways of measuring and comparing results. At present the laboratory data on the absorption of CO_2 laser radiation by the water vapor continuum are, as is well known, are obtained in two ways: 1) with the help of spectrophones and 2) using multipass cells. In spectrophones a small volume of the investigated gaseous medium is studied, and the absorption coefficient is determined with relatively small random measurement errors. The necessity of a preliminary calibration of the apparatus, the methods of doing which are different with different researchers, may, however, lead to noticeable systematic errors. The random errors of the multipass cell measurements are a bit greater, but the systematic errors are reduced to a minimum, since absolute values of the transmissions are measured. In most cases the numerical results of the measurements in the multipass-cell investigations as well can be represented in the form of an absorption coefficient with various forms of dependence on the water vapor partial pressure, the temperature, and the total pressure of the medium. The variety of the forms of these dependences and the different experiment conditions make it difficult to compare the results. For this reason and also taking into account the fact that in the solution of most practical problems it is necessary to know the transmission values, the comparison will be made in terms of transmission values without reducing all the data to the common conditions, in contrast with the approach in Ref. 16, but rather calculating the transmission using Eq. (7) for the specific conditions of each experiment.

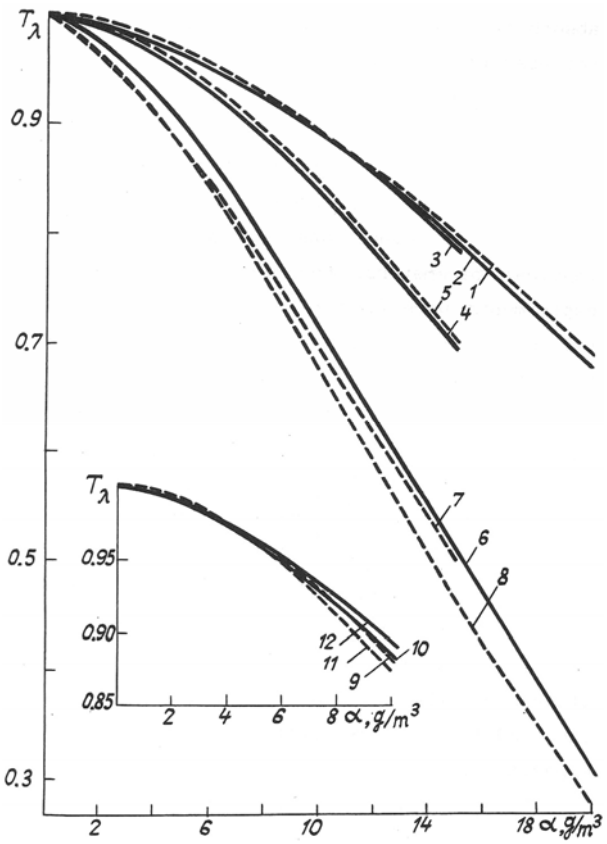

FIG. 5. Transmission of radiation on the line P20 (10.59 μm) of a $^{12}C^{16}O_2$–laser according to the approximations of different authors: 1 and 3) Refs. 12 and 11, respectively, at θ = 293 K for l = 1 km; 5) Ref. 9, 296.5 K, 1.5 km; 7 and 8) Refs. 8 and 24, 300 K, 3 km; 9) Ref. 4, 296 K, 0.98 km; 11) Ref. 10, 300 K, 1 km; 2, 4, 6, 10, and 12) calculations based on Eq. (7) for specific conditions in experiments of all the above-mentioned authors.

The results of calculations of the water vapor transmission of radiation at the CO_2–laser P20 lines (10.6 μm) based on the approximate formulas

of various authors are given in Fig. 5, from which their agreement with the transmission calculations based on Eq. (7) is apparent. The reason for the difference with Ref. 24, from which expression (4) was taken, was indicated above.

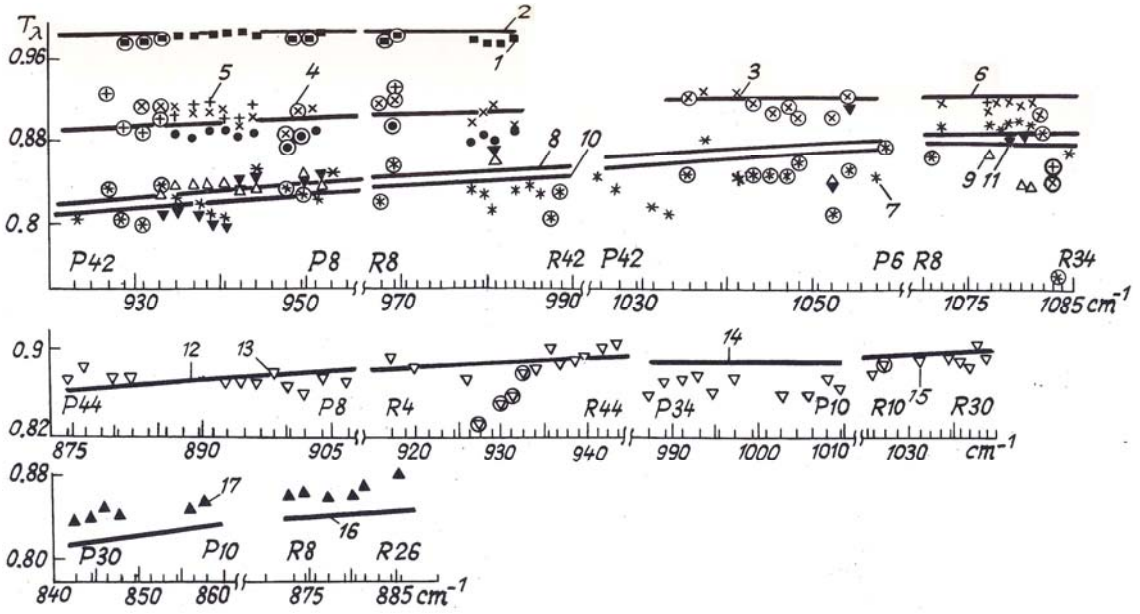

FIG. 6. Transmission of radiation at the line of a CO_2–laser according to the data of different authors: 1) Ref. 10, p = 1.68 Torr, θ = 263 K, L = 1 km; 3, 3, and 5) Refs. 8, 10, and 13, 10 Torr, 300 K, 1 km; 7) Ref. 11, 296 K; 9) Ref. 9, the cell, 295.5 K; 11) Ref. 9, the spectrophone, 298 K at 10 Torr, 1.5 km; 13, 15, and 17) Ref. 11, 9.82, 10.65, and 10.94 Torr, 296 K, 1 km; 2, 6, 8, 10, 12, 14, and 16) calculations based on Eq. (7); 12–15) the $^{13}C^{16}O_2$–laser; 17) the $^{14}C^{16}O_2$–laser, the rest are the $^{12}C^{16}O_2$–laser; 9 and 11) the mixture of water vapor with nitrogen, the rest – moist air at a total pressure of 1 atm.

Figure 6 shows the results of calculations based on Eq. (7) along with the calculations of various authors; measurements for many lines of tunable CO_2–laser. Lines overlapping with the water vapor absorption lines are not considered. Besides, one cannot include in the consideration those lines, ringed in the figure, whose absorption is noticeably affected by ammonia. Even results[8] in which corrections for ammonia have been made do not agree with the calculations as well as the others. The corrections for ammonia in Ref. 8 do not seem to be particularly accurate. A somewhat more noticeable spread and a small systematic shift of the data of Ref. 11 from the calculation is most likely associated with calibration errors of the spectrophone in Ref. 11. On the whole, Fig. 5 shows good agreement between the calculations and the experiments and, consequently, presents the opportunity of using Eq. (7) over the wide spectral range extending all the way from 845 cm^{-1} (≃ 11.8 μm) to 1085 cm^{-1} (≃ 9.2 μm).

Figure 7 demonstrates the possibility of extrapolating expression (7) to the entire 8–13 μm atmospheric window, where calculations based on Eq. (7) agree well with the data of Refs. 5 and 6 for the 8–13 μm region. The stronger absorption in the 8.94 μm microwindow according to Ref. 6 is associated with the possibility of contributions from other absorbing gases (in Ref. 6 a study was made of artificially dried and dampened air) and the lower (2.4 cm^{-1}) resolution, used for the 8.94 μm window, which may be compared with the 0.3–0.5 cm^{-1} for the 11.1, 12.1, and 12.67 μm microwimdows.

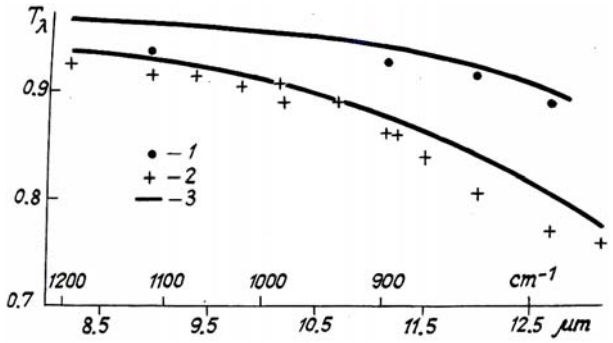

FIG. 7. Transmission of radiation at different wavelengths: 1) Ref 6, moist air at 303 K, 2) Ref. 5, improved data in Ref. 15 at 296 K; 3) calculations based on Eq. (7) at p = 10 Torr, P = 1 atm, L = 1 km and at the temperatures 303 and 296 K.

Figure 8 presents a comparison of calculations based on Eq. (7) with laboratory data[27] obtained

using a diode laser with $\nu = 1203$ cm^{-1} in the temperature range 333–473 K. From Fig. 8 it can be seen that the suggested continuum model qualitatively and satisfactorily quantitatively describes the data of Ref. 27.

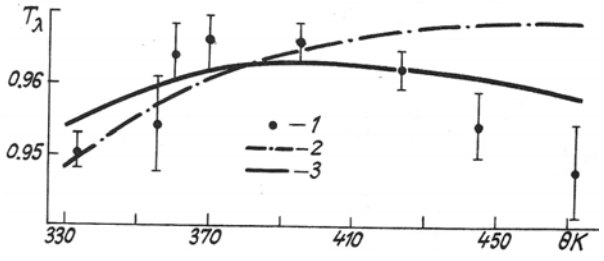

FIG. 8. Transmission of diode laser radiation by water vapor at different temperatures: 1) Ref. 27; 2 and 3) calculations based on Eqs. (2) and (7), respectively, $\nu = 1203$ cm^{-1}, $a = 10$ g/m^3, $L = 1$ km.

Thus, the comparison with the results of the laboratory experiments of various authors (Figs. 5–8) shows that the suggested model of continuous water vapor absorption describes the data obtained by various authors in natural and artificial air over the entire 8–13 μm spectral range at different temperatures.

SELECTIVE WATER VAPOR ABSORPTION

At present it is generally acknowledged that under the conditions of the Earth's troposphere the selective absorption coefficient may be calculated with the help of the Lorentz (dispersion) line shapes:

$$K_\lambda = \frac{S_{ij}\alpha_{ij}}{\pi\cdot[(\nu - \nu_{ij})^2 + \alpha_{ij}^2]}, \quad (8)$$

$$S_{ij} = S_0\left(\frac{\theta_0}{\theta}\right)^m \cdot \exp\left[\frac{E_{ij}}{k\cdot\theta_0}\cdot\frac{\theta - \theta_0}{\theta}\right], \quad (9)$$

$$\alpha_{ij} = \left(\frac{\theta_0^n}{\theta}\right)[\alpha_0(P-p) + \alpha p] = \alpha_0\left(\frac{\theta_0^n}{\theta}\right)[(B-1)p + P], \quad (10)$$

where S_{ij} and S_0, α_{ij} and α_0 are the intensities and the halfwidths at the temperatures θ and θ_0, respectively, and E_{ij} .and ν_{ij} are the energy and the wave number of the lower rotational level and wave number; j is the absorption line of the ith gas; $\nu = 1/\lambda$ is the wave number at which the absorption coefficient k_λ is evaluated; P and p are the total and partial pressures; m and n are temperature parameters; B is the ratio of the self-broadening factor α to the broadening factor α_0, measured at θ_0, and k is the Boltzmann constant.

To evaluate the total absorption coefficient at a specified wavelength (wave number), the expression (8) is summed with respect to i and j, i.e., the gases and their lines that contribute to the absorption at the wavelength λ. Such a method of evaluation (line by line) has gained ground all over the world. To realize it, special atlases (compilations), containing the necessary spectroscopic parameters of the absorption lines,[30–32] have been made. These atlases are continually refined and supplemented with new data. In some cases, however, for laser lines especially, the accuracy of specification of the parameters in these compilations turns out to be insufficient,[33–34] and the necessity arises of their experimental verification.

As of the present time in the 8–13 μm region about 1150 water-vapor absorption lines are known belonging mainly to its purely rotational and fundamental ν_2 bands. The lines are arranged randomly in the spectrum, and some of them coincide with generation lines of CO_2-lasers.

The radiation from such lasers is subjected in the atmosphere to strong absorption equal to the sum of the continuum and selective water vapor absorptions. The mechanisms and behavior of these two types of molecular absorption are different Using the continuum model suggested above, the selective absorption coefficient based on the measured transmission T_λ^e is given by the formulas

$$K_c = \frac{(-\ln T_\lambda^e) - (\ln T_\lambda^K)}{W}, \quad (11)$$

$$W = L\frac{P}{P_0}\cdot\frac{\theta_{00}}{\theta}, \quad (12)$$

where $W_{(\text{atm}\cdot\text{cm})}$ is the water vapor content in the region of the path L (cm), reduced to standard conditions ($P_0 = 1$ atm, $\theta_{00} = 273$ K) and T_λ^K is evaluated according to Eq. (7).

Figure 9 shows the results of measurements of the absorption coefficient of laser radiation on the line $R20$ ($\lambda = 10.25$ μm) of the main carbon dioxide isotope at different partial and the total pressures. The $R20$ line coincides with the water vapor absorption line $13_{-5} \rightarrow 12_{-11}$. The dashed lines in Fig. 9 represent the calculation with the parameters of this absorption line taken from Ref. 30. It is obvious that with continued refinement of the parameters[30] the results of the experiment and calculation draw together. However, complete agreement is not reached, first of all, because the calculation does not take self-broadening into account. Although in the 1986 version[30] a self-broadening factor for some gases is given, it is still absent for the water vapor lines.

Figure 10 shows the results of measurement at the temperatures 293 and 323 K. The temperature dependence of these results was studied in Refs. 10 and 12. In Ref. 10 agreement was obtained with Ref. 30, where $E_{ij} = 1557.85$ cm^{-1}, but in Ref. 12 it was found that $E_{ij} = 1619$ cm^{-1}. Assuming the value of E_{ij} given in Ref. 30 and borrowing the value of the halfwidth of the line α_0 from Ref. 30, we evaluated the rest of the absorption lines parameters $13_{-5} \rightarrow 12_{-11}$ (Table I by the method of least squares from the entire collection of

experimental data), the calculated values of which are shown in Figs. 9 and 10 by the solid lines. (An attempt to fix the value of the line intensity and the distance between the generation and absorption lines led to unreasonable values of L_0, exceeding 0.5–1 cm^{-1}.)

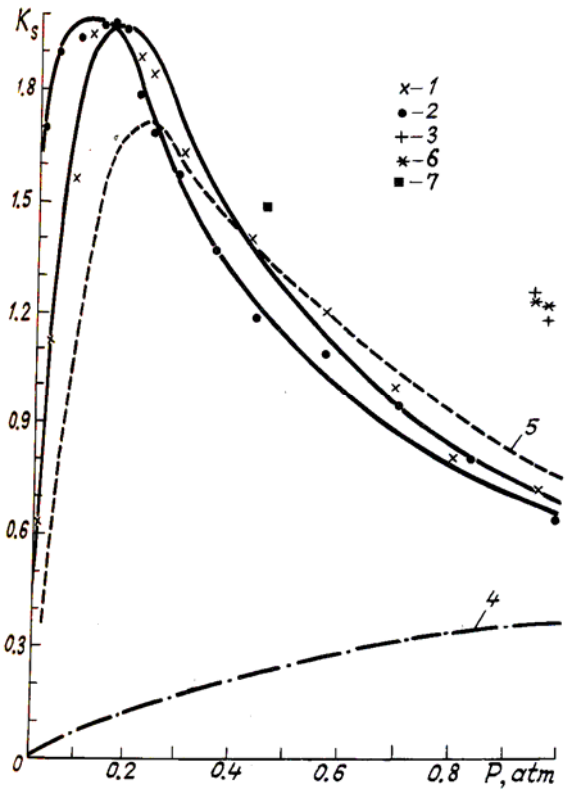

FIG. 9. Absorption of the P20 line of the $^{12}C^{16}O_2$ laser by water vapor at different partial and total pressures: 1 and 2) the experiment at P = 3.82 and 17.3 Torr, 295 K; 4 and 5) calculations using the parameters from Ref. 30; 3) the experiment at p = 10 Torr and 323 K; 6) the calculation at p = 10 and 323 K with the parameters determined in this article; 7) the experiment[35] at 0.01 Torr, 293 K, P = 0.47 atm; $K_s = 10^3 (atm \cdot cm)^{-1}$.

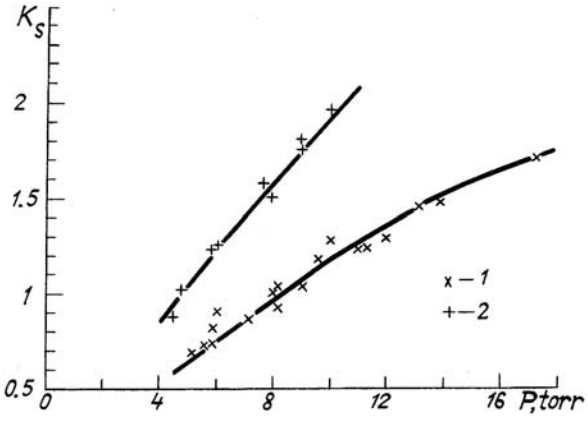

FIG. 10. Absorption of the P20 line of a $^{12}C^{16}O_2$ laser by pure water vapor at different temperatures: 1) 295 K; 2) 323 K; $K_s = 10^3 (atm \cdot cm)^{-1}$.

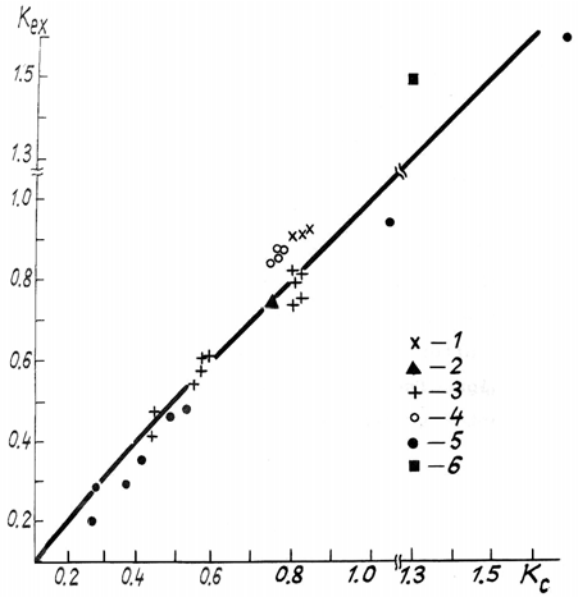

FIG. 11. Comparison of the data of different authors on the absorption of radiation on the P20 line of a $^{12}C^{16}O_2$-laser: 1) Ref. 18, 2) Ref. 9; 3) Ref. 10, 4) Ref. 11, 5) Ref. 12, and 6) Ref. 35.

The published results of absorption coefficient measurements for the $R20$ line (Ref. 8–13) were obtained for different temperatures from 253 to 345 K and partial pressures from a few to a few tens of Torr but with a total pressure of 1 atm (or $\simeq$ 1 atm). Only in Ref. 35 is the absorption coefficient measured at a pressure of 0.47 atm. A comparison of the published data with the results of the present work is given in Fig. 11 in the form of a regression function between the measured (k_e) and calculated (k_c) absorption coefficients (k_c is the sum of the continuum and the selective absorption, calculated according to Eqs. (7) and (8)). As can be seen from Fig. 11, satisfactory agreement is observed on the whole. The differences that do exist between the data of Ref. 35 and the calculated results (see also Fig. 9) must go without comment owing to the absence of any information about the measurement methods in Ref. 35, where only the bare value of the coefficient is given. A more thorough analysis of the data in Fig. 11 shows that the best agreement is observed with the data of Ref. 9, which were obtained with the help of an optical multipass cell. The remaining results were obtained with the help of optical-acoustic spectrophones, while agreeing with the calculations within the limits of the mutual errors, nevertheless indicate the presence of some systematic deviations, since the data of each reference are grouped on one or the other side of the regression line. This systematic is determined by the differences in the calibration of the spectrophones.[13] The value of the random error in the spectrophones measurements random errors is usually not stated by the authors, but one can judge its value from the spread of the

values at constant temperature (see, e.g. , the data of Refs. 8 and 11 in Fig. 11). A similar picture for some other CO_2–laser lines is illustrated by Fig. 12.

Table I also contains parameters of other water vapor absorption lines overlapping with the generation lines of CO_2–lasers. It should be noted, however, that the obtained spectroscopic parameters (the values in the table) of the water vapor absorption lines are used mainly to calculate the absorption of radiation on neighboring laser lines. They may be of insufficient accuracy in other cases because the procedure of finding them requires the a priori knowledge of one of the parameters. The choice of the fixed parameter was to a certain extent arbitrary, even though supported by the tendency toward reasonableness of the obtained values of the parameters in question.

Table I

The parameters of the water vapor absorption lines

Water vapor							CO_2– Laser		
Line, band	ν, cm^{-1}	$S\cdot10^6$, $cm^{-1}(atm \times cm)^{-1}$	$\alpha_0\cdot10^3$, cm^{-1}	$\alpha\cdot10^3$, cm^{-1}	E, cm^{-1}	θ_0	Line, isotope	ν, cm^{-1}	References
1	2	3	4	5	6	7	8	9	10
$13_{-5}\rightarrow12_{-11}$ 000—000	975.9436	145	56.1	—	1557.85	296	R20	—	[30]
	975.9406	126	56.0	250	1557.85	295	$^{12}C^{16}O_2$	924.9740	this paper
$9_5\rightarrow8_{-5}$ 000—000	924.9880	262	46.0	—	885.60	296	P40	—	[30]
	924.9880	210	85.0	340	885.60	295	$^{12}C^{16}O_2$	924.9740	this paper
$8_{-3}\rightarrow9_3$ 010—000	998.8100	56.5	57.0	—	1631.4	296	P22	—	[30]
	998.8054	44.0	56.0	296	1631.4	294	$^{13}C^{16}O_2$	998.7884	this paper
$13_7\rightarrow12_1$ 000—000	970.5705	14.7	36.4	—	2612.8	296	R12	—	[30]
	970.5705	14.6	58.0	284	3663.4	293	$^{12}C^{16}O_2$	970.5472	this paper
$14_3\rightarrow13_{-1}$ 000—000	928.9857	21.5	429	—	2756.42	296		—	[30]
	928.9857	11.1	426	193	2756.42	293	P36	929.0175	this paper
$9_{-2}\rightarrow10_4$ 010—000	928.9550	2.15	535	—	2054.36	296		—	[30]
	928.9550	1.11	532	241	2054.36	293	$^{12}C^{16}O_2$	—	this paper
$9_{-5}\rightarrow10_{-1}$ 010—000	1099.6801	489.6	62.5	—	1718.72	296	R24	—	[30]
	1099.6801	825.0	62.2	351	1718.72	294	$^{12}C^{18}O_2$	1099.4648	this paper
$16_{-3}\rightarrow7_3$ 010—000	1100.9770	1.15	65.4	—	1051.21	296	R26	—	[30]
	1100.9770	1.15	65.1	—	1051.21	294	$^{12}C^{18}O_2$	1100.5928	this paper
$6_{-3}\rightarrow7_3$ 010—000	1101.4510	530	66.3	—	1059.84	296	R28	—	[30]
	1101.4510	740	66.0	388	1059.84	294	$^{12}C^{18}O_2$	1101.7014	this paper
$10_{-7}\rightarrow11_{-5}$ 010—000	1091.2061	562	60	—	1813.22	296	R10	—	[30]
	1091.2061	750	59.7	311	1813.22	294	$^{12}C^{18}O_2$	1091.0247	this paper
$9_{-8}\rightarrow10_{-6}$ 010—000	1074.4091	385	46.5	—	1437.97	296	R14	—	[30]
	1074.4091	841	46.2	223	1437.97	293	$^{12}C^{16}O_2$	1074.6465	this paper
$11_{-7}\rightarrow12_{-3}$ 010—000	1039.4840	675	36.9	—	2275.37	296		—	[30]
	1039.4840	151.3	36.7	171	2275.37	293	P28		this paper
$8_{-2}\rightarrow9_2$ 010—000	1039.5410	28.5	80.0	—	1631.25	296		1039.3694	[30]
	1039.5410	64.2	19.7	341	1631.25	293	$^{12}C^{16}O_2$	—	this paper

RESULTS OF FIELD MEASUREMENTS

Works published as of the present time that contain the results of atmospheric transparency measurements in the 8-13 μm window under field conditions may be conditionally divided into three groups.

In the first group of papers (see, e.g., Ref. 36–38) the authors have tried to process the results of field observations so as to reveal the peculiarities of the continuum absorption. Thus, Ref. 36 and 37 present measurement data on the attenuation of solar radiation by the total thickness of the atmosphere in the 0.65–13.5 μm wavelength range. Dividing the results into groups according to water vapor content, temperature, water vapor pressure and horizontal visibility range makes it

possible to reveal effectively the presence of the temperature dependence and two absorption components in the 8–13 μm window, i.e., the nonlinear dependence of the absorption on the humidity.

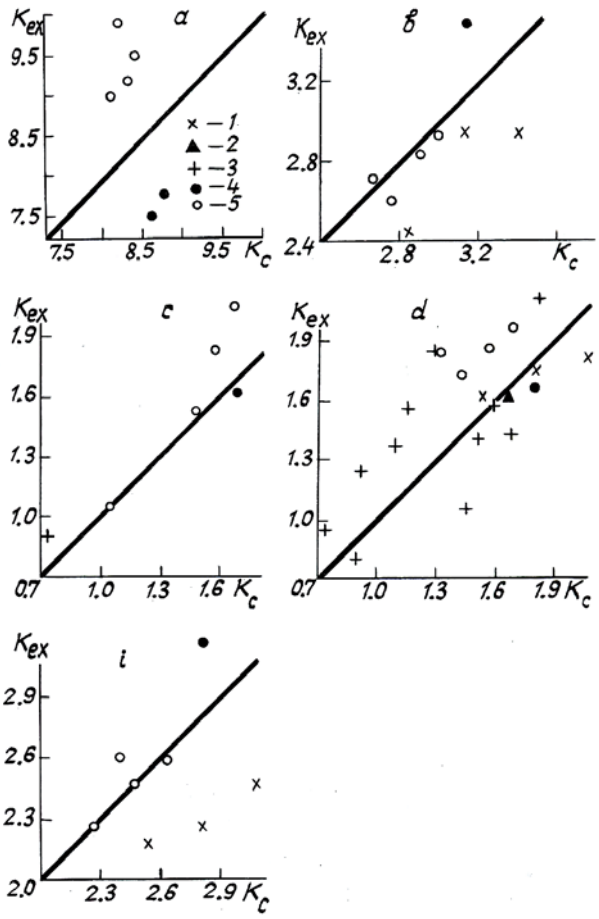

FIG. 12. Comparison of different authors' data on the absorption of radiation on different lines of a $^{12}C^{16}O_2$-laser: a) P40 (10.81 μm); b) R14 (9.31 μm); c) R28 (9.62 μm); 1) Ref. 8; 2) Ref. 9; 3) Ref. 10; 4) Ref. 12; 5) Ref. 11; $K_s = 10^4$ (atm · cm)$^{-1}$.

An analysis was made in Ref. 38 of the relative contribution of aerosol and water vapor dimers and monomers to the radiation extinction on the basis of sky counter-radiation measurements in the 8–13 μm window with a multipass radiometer. It was shown in Ref. 38 that during the warm seasons (the measurements were made between March and November) in the low and mean latitudes the dimer contribution to the absorption prevails.

The second, numerous group of papers (see, e.g., Refs. 39–51) contains the results of field observations, selected, as a rule, on the basis of the minimum possible aerosol contribution. The results were compared either with the laboratory data directly or with calculations based on some model. The general conclusion is that the results agree. The agreement is of a qualitative nature since one usually compares the entire experimental field data file with the approximational dependence obtained from laboratory experiments. The authors of Ref. 47 have suggested a quantitative comparison method, realized later in a number of works (see, e.g., Refs. 48–51) consisting in comparing the experimental results with the results of integrating expression (4) along the ray path with the atmospheric parameters determined simultaneously with each optical measurement. Although expression (4), as was shown in the preceding paragraph, somewhat overestimates the value of the continuum water vapor absorption, the general conclusions of these papers concerning the necessity of accounting in calculations for the real high-altitude profiles of temperature, pressure and humidity in the 8–10 μm region of the molecular absorption of other atmosphere components remain valid.

The third group of papers (see, e.g., Refs. 52–62) is devoted to studies of the aerosol extinction in the region of the 8–13 μm window after the exclusion of the continual absorption from the total extinction. The implemented exclusion methods are various: First, calculations by the approximational laboratory formulas of the continuum absorption and subtraction of it from the total extinction (see, e.g., Ref. 53); second, subtraction of the extinction, measured under especially clear aerosol-free conditions in which the visibility range is more than a few tens of kilometers (see, e.g., Ref. 54); third, separation of the contributions of the various factors by a statistic analysis (see, e. g., Ref. 57); fourth, exclusion of the minimum observed values (see, e.g., Ref. 55).

Both the above-mentioned publications, containing the results of field experiments, the list of which is far from complete, and other papers do not contain all of the specific data needed to carry out the calculations based on model (7). For this reason calculations based on model (7) are compared below with our own measurement data on a horizontal, open multipass path and with the measurement data available to us of the atmospheric solar radiation extinction taken in Antarctica.[49]

It should be recalled that the measurements in the Antarctic were carried out with the help of a field spectral apparatus complex[63] under stable anticyclonal conditions with limitingly low aerosol content, which was monitored by synchronous optical observations of the radiation extinction in the visible spectral range. On the average, this extinction of only 10% exceeded the Rayleigh extinction. It is significant that no correlation was found between the optical widths in the infrared and visible spectral ranges.[49,50]

The regression function between the optical widths τ_r calculated from Eq. (7) using aerological sounding data and the experimentally measured values τ_e (measured in the Antarctic) is given in Fig. 13. For comparison we chose the atmospheric

microwindows in the 10–13 μm region, which are freer of molecular absorption due to other atmospheric components. As can be seen from Fig. 13, the agreement between τ_e and τ_r may be considered to be good. The small excess of τ_e over τ_r, which is about 0.01 and lies, strictly speaking, within the limits of the measurement errors, may be associated with the molecular absorption of other atmospheric components and with the above-mentioned 10% background aerosol extinction.

The air transmission T_e of laser radiation from three carbon dioxide isotopes (about 70 lines) was measured on the horizontal ground-level multipass path in various seasons with the humidity varying from 0.5 to 12 g/m^3, and the temperature from 22 to 26°C. At the same time the meteorological visibility range S_m was monitored and the carbon dioxide concentration, which at the location at which the observations were made was not free from local sources of carbon dioxide, amounted to 340–370 ppm by volume. For the other small atmosphere components, if they affected the absorption of one or another CO_2 laser line, the mean background values of their concentrations were used.[64]

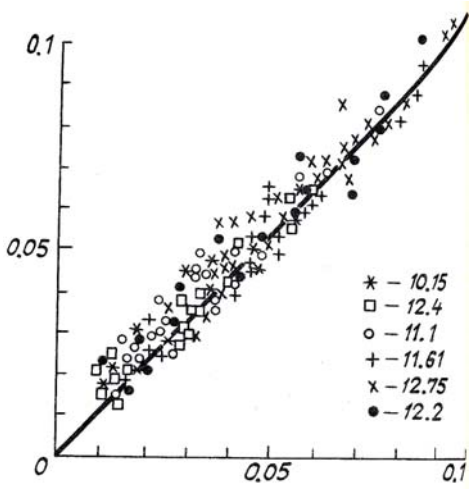

FIG. 13. Comparison of measurement data on the absorption in the 10–13 μm *range by the atmosphere in the Antarctic with the calculations: 1) 10.15; 2) 10.4; 3) 11.1; 4) 11.6; 5) 12.75; 6) 12.2* μm.

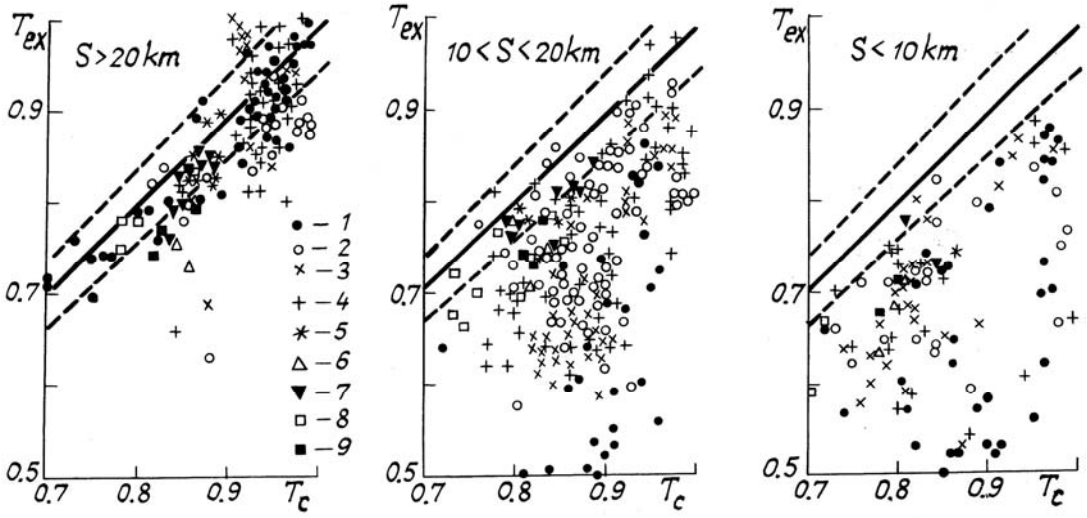

FIG. 14. Comparison of measurements of the transmission of CO_2 laser radiation on a horizontal path with the calculations: 1–4) P20 (10.59), R38 (10.14), R34 (9.20), and P38 (10.79) of a $^{12}C^{14}O_2$ laser; 5, 7) R20 (9.11) and P22 (9.37) of a $^{12}C^{18}O_2$-laser; 6, 8, and 9 R22 (9.67), P24 (11.19), and P24 (10.03) of a $^{13}C^{16}O_2$-laser.

All of the measurements of T_e are grouped into three sets according to the values of the visibility range: $S_m > 20$ km, $10 > S_m > 20$ km, and $S_m < 10$ km. Figure 14 shows the regression functions between T_e and T_r, calculated with the continuous and selective molecular absorption of water vapor and other atmospheric gases at nine lines of CO_2 isotope lasers taken into account. Thus, for example, for the *P*34 $^{12}C^{16}O_2$-laser lines the sulphur dioxide absorption was taken into account, for *P*38 – ammonia, for the other lines of the main isotope - carbon dioxide, etc. The dashed lines mark off the region of the mean square errors. From Fig. 14 it follows that for $S_m > 20$ km, when the role of aerosol is small in the visible region of the spectrum, it is, as a rule, small in the infrared spectral region as well. The observed extinction of CO_2-laser radiation is determined almost completely by molecular absorption and can be evaluated fairly effectively. At the same time, an investigation of the reasons for the occurrence of cases of strong aerosol extinction with a great visibility range remains an interesting scientific problem. For $S_m = 10$–20 km the experimental transmission is m usually less than T_r, i.e., the aerosol contribution to the extinction becomes noticeable. For $S_m < 10$ km the difference between T_e and T_r becomes

still greater and may reach a few tens of a per cent. Here a pronounced seasonal and wavelength dependence of the transmitted radiation was not observed.

CONCLUSION

Our studies of molecular water vapor absorption of radiation in the 8–13 μm atmospheric relative transparency window, performed under field and laboratory conditions, cover a wide range of variation of the parameters affecting the absorption (the total and partial pressure, temperature, humidity, visibility range, and wavelength). Our investigations have confirmed and defined more exactly the main regularities and parameters of the continuum and selective water vapor absorption.

The obtained results have enabled us to formulate an improved semi-empirical continuum model. Calculations using this model agree well with specific results of other works. A comparison of the calculations with the results of field experiments confirmed the important role of the continuum in the extinction of radiation in the 8–13 μm window.

In the case of laser radiation an important role is often played by selective absorption. As a result of our measurements, we have determined the spectroscopic parameters of a number of water vapor absorption lines, overlapping with CO_2-laser generation lines.

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