Studies of 183 GHz water line: broadening and shifting by air, N_2 and O_2 and integral intensity measurements

M.Yu. Tretyakov, V.V. Parshin, M.A. Koshelev, V.N. Shanin,
S.E. Myasnikova, and A.F. Krupnov*

Applied Physics Institute of RAS, 46 Uljanova Street, 603950 Nizhnii Novgorod, Russia

Abstract

The 3_{13}–2_{20} rotational transition of water vapor at 183 GHz was studied using modern resonator spectroscopy methods at atmospheric pressures in the broad frequency range 130–205 GHz down to far wings. The experimental method of sample substitution for the exclusion of the apparatus function was used. The air broadening parameter value was defined as 3.84 ± 0.04 MHz/Torr at 298 K. The observed atmosphere water vapor line center was found to be shifted down at about 53 MHz from the line center at low pressures, which gives a value of -0.07 ± 0.02 MHz/Torr for the air pressure shift parameter. Measurements of broadening and shifting of the water line in pure nitrogen and oxygen atmosphere were also performed. Calculated then parameters of air broadening and shifting agree with directly measured ones within the errors quoted. Measurement of the integral intensity of the line was done. The directly measured integral line intensity coincides with a value given in GEISA and HITRAN databases within experimental error. The results are compared with previous experimental laboratory and satellite data.
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1. Introduction

The 183 GHz water vapor line is one of the most important lines for remote sensing of the atmosphere. Plenty of instruments located on earth surface, high-altitude research aircrafts, balloons, and satellites used for the atmosphere studies and weather prediction are utilizing the 183 GHz line in particular for determination of the water content. Results of the measurements from instruments are then used for recovery of atmospheric parameters using the spectral parameters of the line obtained in laboratory experiments as one of the most important inputs. The accuracy of the recovered atmospheric parameters directly depends on the accuracy of laboratory measurements of parameters of the line.

Until recently only the line intensity and the line pressure broadening parameters were used in practice of remote sensing. It was known, though, that there is another intrinsic spectral line parameter—pressure line shift. Pressure line shift was not used in remote sensing practice because on one side there was no clear manifestation of this effect in remote sensing data, and on another side there were no laboratory data showing the existence of an appreciable air pressure shift of the line.

Authors of [1] found an air pressure shift of the 183 GHz water vapor line by analyzing MLS and MAS satellite limb sounding data. The very first results showed improvement in the fitting of the satellite data several times by introducing the 183 GHz water line air pressure shift into the calculation.

As it was shown in [1], the value of the line shift parameter as well as line broadening parameter can be obtained by fitting limb sounder data. Nevertheless, if the parameters can be known a priori, from laboratory experiment, this reduces the number of adjustable parameters in the model and considerably increases the accuracy of the recovery of other atmospheric parameters.

The accurate laboratory measurements of the line spectroscopic parameters are also important for development of a new, more accurate, atmospheric radiation transfer model.

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* Corresponding author. Fax: +7-8312-36-37-92.
E-mail address: kru@appl.sci-nnov.ru (A.F. Krupnov).
URL: http://www.appl.sci-nnov.ru/mwl.

The water vapor line at 183 GHz was also studied by a number of investigators both in real atmosphere [2,3] and in laboratory at low [4,5] and high [6,7] pressures, but the line center pressure shift was not ever mentioned by them. Moreover, the line pressure broadening parameter values obtained by different authors do not quite coincide even within experimental errors given by authors.

The goals of the present investigation were first of all to measure the shifting parameter of the water line center by pressure of air in laboratory experiment, second to refine the value of the line pressure broadening parameter, and third to measure the line integral intensity.

2. Experimental details and results

Millimeter-wave resonator spectrometer with fast digital frequency scanning described in detail in [7] was used. The principle of radiation absorption measurement by the apparatus consists in the measurement of width of resonance curve of resonator at the set of frequency points within required range. The spectrometer employs synthesized radiation source based on Backward Wave Oscillator and open Fabry–Perot resonator as the radiation absorption sensor. The measurement of resonance width (which depends on radiation absorption in a sample placed in the resonator) was done at any experimental frequency point by recording the resonance curve and its following fitting to a theoretical model (Lorentz curve). Averaging of results of fitting of about 300 curves recorded during back and forth fast digital radiation frequency scans was performed at every frequency point to increase the resonance width measurement accuracy.

Broadband records in the range 130–205 GHz were obtained by jumping from one longitudinal mode of the resonator to another one without changing the resonator length. The effective resonator length was determined from known radiation frequency and assumption of integer number of the half-waves in between mirrors with sub-micron accuracy and eigen-frequencies of the resonator were calculated. The resonator length was near 400 mm, so the inter-mode spacing was about 380 MHz. To get a compromise between minimizing the experiment time and getting the informative water line record the resonator modes thinning was used at wings of the line: every 8th longitudinal mode was selected in the range 130–150 GHz, 4th in 150–165 GHz, 2nd in 165–175 and in 190–205 GHz, and every next mode in the 175–190 GHz range. The total set of resonator eigen-frequencies selected to be used as experimental frequency points contained about 90 numbers.

For the present study plastic film isolation was made for the Fabry–Perot resonator to minimize the effect of

airflows and variations of ambient atmosphere conditions. The isolation allowed us to fill the resonator not only by laboratory air but also by pure nitrogen and oxygen. The sample humidity and temperature monitoring in course of experiment was performed by "TESTO-645" [8] equipped with highly accurate humidity/temperature probe. Guaranteed accuracy of relative humidity measurement is $\pm 1\%$ (10–90% RH, from +15 to +30 °C) and $\pm 2\%$ in the remaining range. The accuracy of temperature measurement is ± 0.2 °C. Pressure meter (600–800 mmHg range), calibrated with an accuracy of 0.5 mmHg by the state metrology service, was used for the sample pressure measurement.

As it is known, the width of the Fabry–Perot resonance can be written as

$$\Delta f = \frac{cP_{\text{total}}}{2\pi L}, \tag{1}$$

where c is the light velocity in a substance, L is the Fabry–Perot resonator length, and P_{total} are the total relative losses of radiation energy during one traversing of the resonator. Total losses P_{total} in its turn consist of radiation losses in the resonator itself and losses (absorption) in the gaseous sample filling the resonator:

$$P_{\text{total}} = P_{\text{reflection}} + P_{\text{coupling}} + P_{\text{diffraction}} + P_{\text{sample}}$$
$$= P_{\text{resonator}} + P_{\text{sample}}. \tag{2}$$

More details about losses in resonator are given in [9]. The sample losses P_{sample} can be expressed through the resonator length L and absorption coefficient γ using Lambert's law as

$$P_{\text{sample}} = 1 - \exp(-\gamma L), \quad \text{or}$$
$$P_{\text{sample}} = \gamma L \text{ (for } \gamma L \ll 1\text{).} \tag{3}$$

Thus, to calculate the absolute value of the sample absorption coefficient γ from the measured width of resonance Δf one needs to determine the resonator losses $P_{\text{resonator}}$. In our earlier studies we used for this reason either calculated values of resonator losses [7] or method of the resonator length variation [10]. Having possibility to fill the resonator by non-absorbing substance (e.g., pure nitrogen) and measuring the resonance width Δf_0, we were able to get the resonator losses just from experiment:

$$P_{\text{resonator}} = \frac{2\pi L \Delta f_0}{c}. \tag{4}$$

Then the sample absorption coefficient γ can be easily obtained from Eqs. (1)–(4) and it is especially simple in case of $\gamma L \ll 1$ (which really corresponds to our case):

$$\gamma = \frac{2\pi}{c} \cdot (\Delta f - \Delta f_0). \tag{5}$$

To fill the resonator with non-absorbing sample we used in the experiment a permanent flow of pure nitrogen evaporated from a liquid phase and heated up to the

room temperature. The nitrogen replaced gradually an air inside our resonator isolation and forced out water molecules from the isolation walls and parts of the resonator, so in a few hours we were able to get zero humidity indication from "TESTO-645," whose probe was located nearby an inter-mirror space of the Fabry–Perot resonator. The resonator losses obtained as described above through the measurement of the resonance width Δf_0 at every one from the set of resonator eigen-frequencies in such non-absorbing atmosphere conditions gave us an apparatus function $P_{\text{resonator}}$. The function is shown in Fig. 1 as the trace *B*.

As it was assumed, the function increases with frequency with small irregular variations. These variations correspond by our opinion to the remaining radiation interference in the apparatus horns and wave guides and are related to the small change of the angle of radiation beam exciting the resonator and corresponding change of the coupling losses with frequency. In addition to that, precise analysis of the function Trace *B* showed up the presence of a residual weak spectral line at 183 GHz, corresponding to about 0.4% relative humidity of the sample (or 0.1 g/m^3 of remaining water content) at zero humidity indication by "TESTO-645," having 2% guaranteed accuracy. Corresponding correction for the apparatus function was calculated (trace *C* in Fig. 1) using MPM-93 program [11]. Apparatus function then was obtained by subtraction of residual water absorption from Trace *B*.

The apparatus function turned out to be very reproducible. Four traces obtained one after another in two days between each experiment had a noise-like difference with a standard deviation less than 0.2% from the average amplitude.
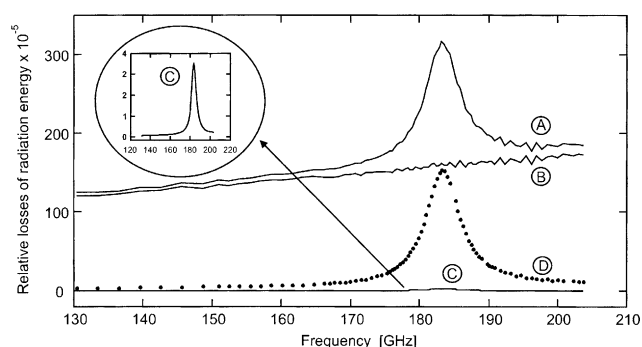

Fig. 1. Obtained from the experiment total relative losses of radiation energy during one traversing of the Fabry–Perot resonator filled by laboratory air P_{total} at temperature 24.5 °C, pressure 755.8 mmHg, and humidity 4.75 g/m^3 (trace *A*). Obtained from experiment the losses of radiation in the resonator through which the flow of nitrogen at the same temperature was maintained (trace *B*). Calculated losses of radiation due to residual water in the resonator while maintaining nitrogen flow (trace *C* and also zoomed in interposed picture). The radiation losses in the laboratory air P_{sample} obtained as $A - (B - C)$ (trace *D*).

The trace *A* in Fig. 1 presents total losses of the Fabry–Perot resonator P_{total} filled by laboratory air, at ordinary room pressure, temperature and humidity. The trace was obtained by measurement of resonance width Δf using the same procedure as that for the apparatus function at exactly the same frequencies from the aforementioned set. The minor change of effective resonator length (about 20 μm) due to different refractive indexes of laboratory air and pure nitrogen was compensated by mechanical change of the resonator length and taken into consideration.

The trace *D* in Fig. 1 presents the value of the relative radiation losses in laboratory air P_{sample} (which is in the fact spectrum of atmospheric absorption) under given experimental conditions calculated in accordance with Eq. (2). for each of the eigen-frequencies from the set. The trace is shown in points to give an idea about the location of experimental frequencies in all of traces of Fig. 1.

As it is known, the spectrum of atmospheric absorption in this range consists of the single water vapor line located on non-linear slope of summed wings of other higher frequency water lines and continuum absorption spectrum (see e.g., [11]). So, to extract spectroscopic parameters of the observed line and separate the absorption in the line and non-resonance absorption, we fitted the record of absorption obtained from the experiment to the Van Vleck–Weisskopf profile (which was selected in our earlier studies [7,10] as that best suitable for atmospheric lines) with addition of linear, quadratic, and constant with frequency terms:

$$F(v) = a \cdot v^2 \cdot \left(\frac{\Delta v}{(\Delta v)^2 + (v - v_0)^2} + \frac{\Delta v}{(\Delta v)^2 + (v + v_0)^2} \right)$$
$$+ b \cdot v^2 + c \cdot v + d,$$

where a, Δv, and v_0 are the line amplitude, half width and the line center correspondingly; b, c, and d are non-resonance absorption characterizing constants. All of these six constants were obtained from the fit. No systematical variation of the observed line shape from the fitting model exceeding experimental noise was observed.

Leaving the analysis of non-resonance absorption for the future studies we concentrated on the line parameters, which were extracted from the observed line shape.

Dry air broadening parameter Δv_b^{air} was calculated from the obtained line width using the formula:

$$\Delta v = \Delta v_b^{\text{air}} \cdot (p_{\text{atm}} - p_{\text{water}}) + \Delta v_b^{\text{water}} \cdot p_{\text{water}},$$

where Δv is the line half width obtained from Van Vleck–Weisskopf profile fit; p_{atm} is the atmosphere pressure at the time of experiment (pressure of the air sample in the resonator isolation); p_{water} is the partial pressure of water vapor in the air sample, which can be calculated from measured humidity and temperature;

$\Delta v_b^{\text{water}}$ is the broadening parameter of the line for pure water vapor, which was measured by us earlier [12] as 21.69(45) MHz/Torr at a temperature of 296.8 K.

In all our experiments we found that the line center frequency v_0 obtained from the fit was lower than that measured at low pressures (e.g., $v_{\text{line}} = 183\ 310.0906$ MHz, obtained from beam measurements [13]). If one interprets this effect as due to the pressure shift, then the pressure shift parameter for this line can be estimated using expression:

$$\Delta v_s = \frac{(v_{\text{line}} - v_0)}{p_{\text{atm}}}.$$

The experiment was repeated four times within one-week period to get an idea about the possible systematic errors. The results are presented in Table 1. Statistical errors presenting standard deviation of parameters determined from fit (arising basically from the observed atmospheric water line signal to noise ratio) are given in the table. As it may be seen, the variations of the parameter values due to some other uncontrolled factors are about five times larger than the statistical error.

The broadening parameters from all experiments were recalculated to the temperature 298 K (for further averaging) using common expression

$$\Delta v_b(T) = \Delta v_b(T_0) \cdot \left(\frac{T_0}{T}\right)^n \tag{6}$$

and value of $n = 0.64$ was experimentally obtained for air broadening of this line in [14]. Subsequent averaging of obtained by such way data gave the air broadening parameter value as 3.84 (4) MHz/Torr, where the given error is doubled standard deviation of these four data.

Then we measured the water line broadening and shifting parameters by two major atmospheric components by use of the same method filling the Fabry–Perot resonator by pure nitrogen and oxygen both enriched by water vapor. The permanent gas flow through the resonator isolation was used as in the case of the apparatus function measurements described above. Technically pure (volume part of about 98%) gases from standard high-pressure gas-cylinders were used. The gas was flowed through the closed bulb with liquid pure water to enrich the sample by water vapor. In a few hours of flowing we were able to get satisfactory stable sample conditions inside the resonator isolation. The sample humidity in both experiments was near 3 g/m³.

Absorption of radiation in oxygen line at 118.7 GHz was used as additional sample compound control in both nitrogen and oxygen experiments. In the nitrogen experiment no additional absorption was observed at 118.7 GHz in comparison with the apparatus function measurements. Small regular absorption increasing at 118.7 GHz due to water vapor presence in the sample was calculated and taken into account using MPM program [11] as it was described above. In the oxygen experiment near five times higher absorption at 118.7 GHz in comparison with absorption in air was observed, which corresponded to about 98% of oxygen content inside our isolation.

The apparatus function was taken separately for every measurement using the same methodics described above.

Obtained data for the line pressure broadening and shifting by nitrogen and oxygen parameters are presented in Table 2 together with air broadening and shifting for averaged air conditions of experiments described above. Uncertainty of parameters given in brackets for nitrogen and oxygen experiments correspond to doubled (to take into account possible systematical errors) statistical error of the parameters determination in one experiment.

Data from the table allow us to compare the value of the dry air broadening parameter directly measured in the air experiment with value of the parameter calculated from nitrogen and oxygen experiments using known 79/21 percent contributions of the gases in air. The temperature dependences of the parameters can be neglected in this case since the temperature differences between experiments are within 1°. Calculated value of the broadening parameter

$$\Delta v_b^{\text{air\_calc}} = 0.79 \cdot \Delta v_b^{\text{nitrogen}} + 0.21 \cdot \Delta v_b^{\text{oxygen}}$$
$$= 3.89\ \text{MHz/Torr}$$

Table 1
Experimentally obtained pressure broadening and shifting parameters for the 183 GHz water vapor line in air

Dry air pressure broadening parameter (Δv_b^{air}), MHz/Torr	Air pressure shift parameter (Δv_s), kHz/Torr	Temperature, K
3.872 (13)	−78.0 (66)	295.9
3.870 (12)	−66.0 (53)	297.4
3.831 (12)	−70.0 (56)	297.6
3.804 (12)	−56.7 (54)	299.2

Statistical error of parameter determination is given in brackets in units of the last given digit of the parameter.

Table 2
Experimental pressure broadening and shifting parameters for the water vapor line at 183 GHz in atmosphere of different gases

	Broadening parameter, MHz/Torr	Shift parameter, kHz/Torr	Temperature, K
H_2O in nitrogen	4.24 (4)	−91 (24)	299
H_2O in oxygen	2.57 (5)	−84 (30)	298.6
H_2O in air[a]	3.84 (4)	−67 (16)	298

[a] This value was obtained by averaging of data from Table 1.

coincides within given experimental uncertainties with the averaged experimental value given in Table 2. Similar conclusion can be made regarding the line shift parameters.

Another experiment was undertaken to investigate integral intensity measurements by the methodics described and also to check consistence of the experimental method. The water line was recorded seven times under sequentially decreasing sample humidity by the gradual replacement of the air inside the resonator isolation by pure nitrogen. Obtained records are presented in Fig. 2. Then the observed line widths and amplitudes were obtained as described above and integral line intensities were calculated as the line amplitude (the line absorption coefficient at the line central frequency) multiplied by the π times line half width ($\pi \cdot \gamma(v_0) \cdot \Delta v$) for each experiment. Gas temperature inside the resonator in course of all experiments was kept near 297 K. The integral intensity points plotted in Fig. 3 vs. measured absolute humidity of the sample demonstrate very well linear dependence. The line has practically zero intercept, as it should be in accordance with theory. Value of the normalized line intensity by concentration

of water molecules (N) calculated from these data as $\pi \cdot \gamma(v_0) \cdot \Delta v/N$ averaged over all seven measurements and was expressed in units of cm/molecule constitutes $0.780(8) \times 10^{-22}$, where the given error corresponds to standard deviation of these seven numbers. For comparison GEISA [15] and HITRAN [16] databanks for 296 K present a value of 0.777×10^{-22} cm/molecule. Thus, the line intensity directly measured in our experiment coincides with the tabulated one within 0.4%.

This experiment demonstrates that the sample absolute humidity can be measured in such experiment with high accuracy using tabulated value of the line intensity, which may lead to many practical applications.

3. Discussion

Except accurate measurement of atmospheric line parameters, our broadband laboratory measurements of atmospheric absorption allow a rigorous check of atmospheric radiation absorption models widely used in remote atmosphere sensing and other applications. Comparison of our data with MPM model [11] is given in Fig. 4. Points represent the experimental trace, while the calculated one is shown by solid line. Experimental values of the air pressure and temperature were used for the model calculation, while the air humidity was varied a little around measured value within the humidity-meter accuracy (1% RH). The best agreement between experiment and model was achieved when decreased in 0.4% RH (or 0.1 g/m^3) humidity (as compared to the measured by humidity meter) was used for the model trace calculation. Similar variation of atmospheric pressure and temperature within measurement accuracy limits did not show any essential effect.

Enlarged difference between experiment and calculation is shown in lower part of the Fig. 4. It demonstrates
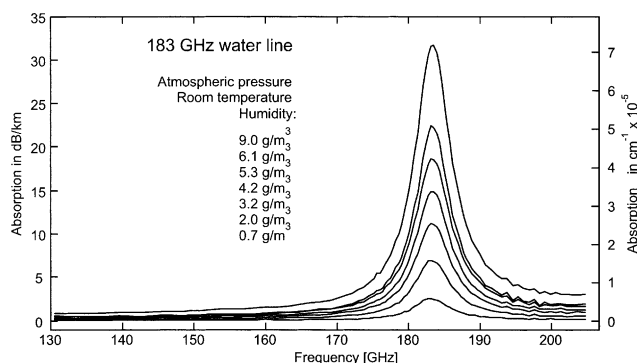

Fig. 2. Absorption spectrum of a mixture of laboratory air and dry nitrogen around the water line at 183 GHz at atmospheric pressure and room temperature recorded under different humidities (shown in the figure).
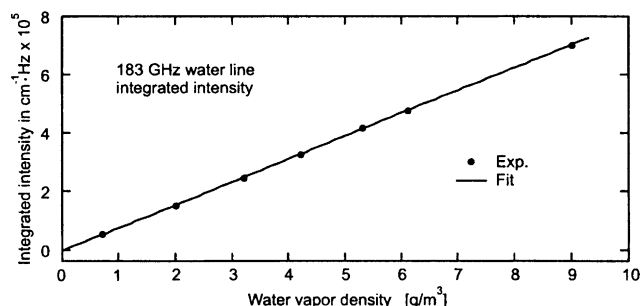

Fig. 3. Experimentally measured 183 GHz water line integral intensity vs. humidity of mixture of laboratory air and dry nitrogen situated inside the resonator (points) and linear approximation of the experimental dependence (solid line).
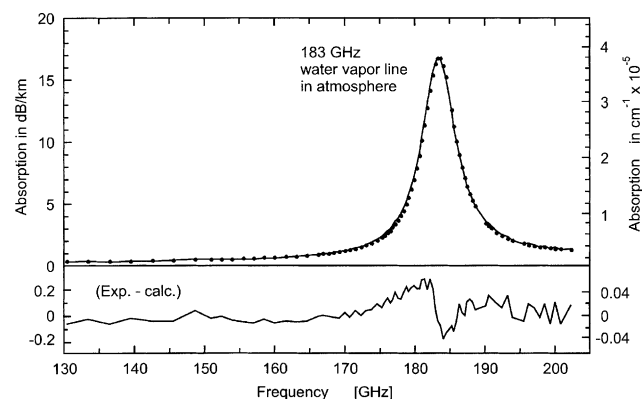

Fig. 4. Comparison of experimental absorption (points) with the calculated one (solid line in upper trace) using MPM program. Enlarged difference is shown in lower part by solid line. Atmospheric parameters used for calculation: pressure 756 mmHg; temperature 24.5 °C; and humidity 4.65 g/m^3.

very good general coincidence of both traces. Maximal value of difference is reached near the water line center but it constitutes less than 2% from the absorption value. The systematic character of the difference within the 2% mentioned demonstrates both—the presence of the pressure line shift lacking in the MPM model and slightly different line widths used in MPM model and obtained in our experiment.

The review of the 183 GHz water line air broadening parameter values obtained by different authors before year 2000 was done in [1]. The comparison of the value obtained from our experiment with these data is given in Fig. 5. Data for the figure are taken from [1], not from original publications. Our points (from [7] and the present work) were recalculated to 300 K using Eq. (6).

Obtained in this work the dry air pressure broadening parameter for the 183 GHz water line is also very close to the calculated by R. Gamache value of 3.96 MHz/Torr, which was mentioned in [1] as personal communication, 1997.

The air pressure shift of the 183 GHz water transition was reported in [1] where it was obtained by fitting data from two different microwave limb-sounding instruments. Two slightly different negative values of the pressure shift parameter -0.19 ± 0.04 and -0.27 ± 0.05 MHz/Torr were presented in [1]. In the same paper authors refer to the calculated by R. Gamache value -0.13 MHz/Torr which is cited as personal communication in 1997.

In our experiment for the first time the pressure shift parameter for 183 GHz water line was observed in the

laboratory. Direct observation of the water vapor line profile in the air at atmospheric pressure and that usual for laboratory molecular spectroscopy procedure of line shape fitting were used. Our experimental value of air pressure shift parameter was obtained as -0.07 ± 0.02 MHz/Torr. As far as we know, this is the first case when the same atmospheric line pressure shift measurements were taken independently both from satellite and laboratory measurements. Comparison of the results of them demonstrates, by our opinion, a good agreement, both in the sign and order of magnitude of the air shift parameter, having in mind all differences in experiments and recovery of the data. In particular, limb sounding assumes that signal from the spectral line is formed in different layers of atmosphere on the line of sight differing by, e.g., temperature; but for recovery of the shifting parameter the knowledge of its temperature dependence is needed. Most reliable could be experimental temperature dependence, which is absent at the moment for the shift. Also good agreement by our opinion is shown to the calculated value of the shift.

4. Conclusions

Improvements of the experimental technique achieved in [7,10] and in the present work permitted accurate analysis of the 183 GHz water line in laboratory air as well as in pure nitrogen and oxygen at atmospheric pressure and room temperature. Direct measurement of apparatus function in the experiment and subtracting it from the observed spectrum allowed a several times increase of the signal to noise ratio of the observed atmospheric water line compared to our previous experiments. Refined values of the line broadening parameters by pressure of air, nitrogen and oxygen were obtained. Our laboratory study confirmed the presence, sign, and order of magnitude of air pressure shifting parameter for this line obtained for the first time from analysis of limb sounding data. It was demonstrated that our measurement method allowed also a direct measurement of the spectral line integral intensity. Measured integral intensity for the water line well coincides with the value given in GEISA and HITRAN spectroscopic databanks. Spectroscopic parameters of the water line obtained in the present work can be used for the atmosphere remote sensing and also for refinement of existent atmospheric radiation transfer models.
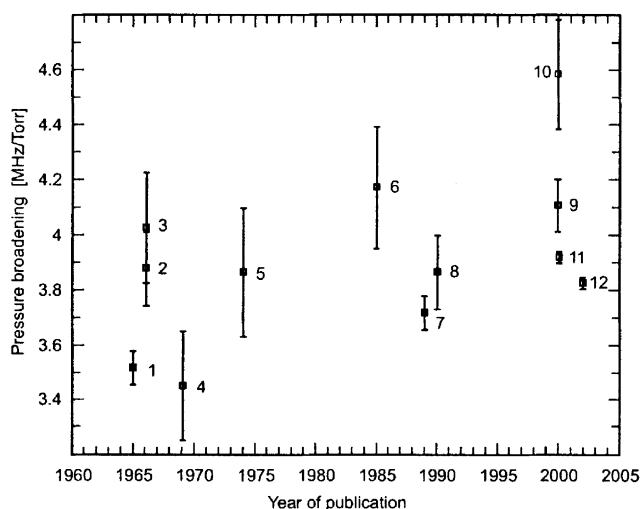

Fig. 5. Measured values of the water line air pressure broadening parameter. The authors responsible are as follows. (1) Rusk [5]; (2) Dryagin et al. [3]; (3) Frenkel and Woods [6]; (4) Hemmi and Straiton [17]; (5) Ryadov and Furashov [2]; (6) Bauer et al. [4]; (7) Bauer et al. [14]; (8) Goyette and DeLucia [18]; (9) Pumphrey and Buehler [1]—MLS; (10) Pumphrey and Buehler [1]—MAS; (11) Krupnov et al. [7] (obtained using the calculated apparatus function); (12) This work (obtained using the measured apparatus function).

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