

Medium resolution transmission measurements of water vapor at high temperature

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Abstract

Medium resolution transmissivities of water vapor were measured at temperatures between 600 K and 1550 K for all important vibration-rotation bands as well as part of the purely rotational band. Measurements were made with an improved drop tube design, which ensures a relatively isothermal high-temperature gas column. Data were collected with an FTIR-spectrometer, allowing for much better spectral resolution than most previous high-temperature measurements. The measured data were compared with the HITEMP database, as well as with the data of Phillips for the $2.7 \mu\text{m}$ band of H_2O . The data show minor discrepancies with the high-resolution database, particularly at higher temperatures, but in general agreement is acceptable.

1 INTRODUCTION

Knowledge of radiative properties of combustion gases is required to accurately predict radiative fluxes in a number of physical systems like fires and combustion systems. Unfortunately, absorption coefficients of absorbing gases are not known with sufficient accuracy to make reliable heat transfer calculations, especially at high temperatures. Spectra of gases broadened by N_2 , air and other buffer gases have been investigated by a number of researchers. For example, Rinsland et al. [1] describe atmospheric measurements of water vapor absorption using a telescope and an FTIR spectrometer. Atmospheric measurements have also been made with FTIR spectrometers for ozone (Bouazza et al. [2] and Flaud et al. [3]) and CO (Farrenq et al. [4]). Atmospheric measurements permit long optical paths, at the cost of isothermality and homogeneity of the path. And, of course, it is not possible to make high-temperature atmospheric measurements.

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Laboratory measurements are made with window cells (uncooled [5–7], or water-cooled [8–10]), nozzle seal cells [11–14] and free jet devices [15, 16]. Hot-window cells consist of a gas column confined within a container by windows at the ends. While isothermality of the gas is ensured, such measurements are usually limited to temperatures of below 800 K [17]. Cold window cells use water-cooled windows, thus allowing higher temperatures. However, it is impossible to obtain a truly isothermal gas column with such a device [8]. Nozzle seal cells contain the combustion gases within open flow cells by means of layers of inert gases (N_2 or Argon) on either end. While these cells eliminate some of the problems associated with windows, they may cause density and temperature gradients near the seal, as well as scattering by the turbulent eddies of the mixing flows [18]. Free jet devices use a burner and jet, and may thus be used for extremely high temperatures, at the cost of considerable uncertainty in the gas temperature and density distribution, as well as the path length.

Radiation measurements have been made for various water vapor bands by other researchers. Esplin et al. [19] have measured line positions and line intensities for lines in the $720\text{--}1400\text{ cm}^{-1}$ spectral region of H_2O . Their measurements were made at 20 Torr and 1000 K, in a heated 1.75 m single pass absorption cell. Phillips [20] has made moderately high resolution measurements (0.06 cm^{-1}) for the $2.7\text{ }\mu\text{m}$ band of H_2O at temperatures of up to a 1000 K, which were later degraded to 25 cm^{-1} . These measurements were made with a Nicolet 8000 FTIR spectrometer equipped with a CaF_2 beam splitter and a globar source. The data were obtained over a range of optical depths from 0.4 to 20.3 atm-cm. Band-model parameters were obtained from the averaged data assuming a combined Lorentz-Doppler model, and comparisons were made with other band-model data and with line-by-line calculations.

Flechl et al. [21] have made comparisons of experimental and HITEMP [22] transmissivities for CO, CO_2 and H_2O for temperatures of up to 1100°C . Their measurements were made at a resolution of 0.5 cm^{-1} , and required subtraction of the emission signal from the total of emission and reflection. Their results for CO_2 show higher absorption than HITEMP at 1100° in the wings of the $4.3\text{ }\mu\text{m}$ band, similar to our earlier CO_2 measurements [17, 23]. Flechl’s high-temperature data for H_2O show higher absorption than HITEMP for some parts of the $6\text{ }\mu\text{m}$ band, and less absorption than HITEMP for other parts of the same band, similar to the data presented below. In some cases, their measured absorptances are as much as 50% off from HITEMP predictions.

In the present work, medium resolution (4 cm^{-1}) transmission measurements were made for the rotational, $6.3\text{ }\mu\text{m}$, $2.7\text{ }\mu\text{m}$ and $1.8\text{ }\mu\text{m}$ bands of H_2O with a drop tube mechanism and FTIR spectrometer. These measurements were made at a total pressure of 1 atm, for $\text{H}_2\text{O}/\text{N}_2$ mixtures at temperatures of up to 1550 K. The setup used for our earlier CO_2 measurements [17, 23] was modified for these measurements. These modifications consisted of opening up the optical path (using larger mirrors and openings) so as to allow a better signal, as well as switching the locations of the source and the detector [24] to eliminate furnace emission. Remodulated furnace emission was eliminated by taking advantage of the cube-corner optics within the FTIR spectrometer, as described in [24].

These measurements were compared with transmissivities obtained from the HITEMP database [22]. This database lists line locations, strengths and half-widths for various gases. These line data were used to obtain narrow-band transmissivities, using a custom-written code. For further details of the code, as well as of the procedure adopted to obtain transmission data from HITEMP, the reader is referred to [17, 23].

2 EXPERIMENTAL DETAILS

Narrow-band H₂O transmission measurements were made at temperatures of up to 1550K with a resolution of 4 cm⁻¹ using a drop tube mechanism and FTIR spectrometer. A diagram of the setup is shown in Figure 1.

The setup used for our earlier CO₂ measurements [17, 23] was modified so as to (i) allow a larger signal (ii) eliminate modulated furnace emission. As Figure 1 shows, steam from a steam generator was mixed with preheated N₂. The mole fraction of H₂O was varied by varying the flow rates of N₂ and steam. An Agilent series Micro Gas Chromatograph was used to measure the mole fraction of H₂O within the furnace. A ZnSe window at the bottom of the drop tube prevented the absorbing mixture from entering the rest of the optical path, which was purged with dry N₂ to eliminate atmospheric CO₂ and H₂O. Further details of the setup may be found in [23] and [17].

The transmissivity of H₂O at a given temperature and pressure path length was obtained by ratioing the signal obtained with the required concentration of H₂O at that temperature with the signal obtained without H₂O (i.e., with the furnace filled with N₂) at the same temperature. Six spectra were taken with 0% H₂O at each length and temperature, and these were averaged to give the base signal at that length and temperature. Another six spectra were taken at each measurement condition (length, temperature and H₂O mole fraction), and these spectra were ratioed with the base spectra at that length and temperature to obtain the transmissivity for that measurement condition. The six transmission spectra were then used to calculate an average narrow-band transmissivity, as well as a standard deviation as an estimate of the random error of the measurement.

Since the modulated emission from the furnace was eliminated [24], it was no longer necessary to subtract the emission signal from the total of the emission and reflection as described in [17]. Data were collected using a wide-band MCT detector with a KBr beamsplitter and an IR source for the rotational, 6.3 μm and 2.7 μm bands of H₂O, and an InGaAs detector with a Quartz beamsplitter and tungsten-halogen source for the 1.8 μm band.

3 DATA ANALYSIS

The measured data at each temperature are compared with results from the HITEMP database. The linelist calculated for a nominal 1000 K for water vapor was used to make the comparisons. Areas of differences and agreement with the database are identified. Line data from the HITEMP database are used to calculate narrow band transmissivities at the measured resolution, taking into account instrument broadening by the FTIR spectrometer. The spectrometer used triangular apodization with a half-width at the base of 4 cm⁻¹, and thus a sinc² function was used to model the instrument broadening, as explained in [23]. The measurements were made over a period of 8 to 12 hours for each temperature, which made it necessary to correct for the drift in the strength of the signal at the detector. This drift could be due to temperature fluctuations of the optical components (mirrors and beamsplitters) or varying detectivity of the cryogenic MCT detector. Since the transmissivity was obtained as the ratio of two signals, the ‘baseline transmissivity,’ which is the transmissivity in the regions of the spectrum over which the gas does not absorb radiation, is expected to be 100%. However, since the signal may be varying with time, the baseline transmissivity could be different from 100% in these regions. Thus, an estimate of the factor by which the signal has changed at each spectral location within the bands may be obtained by

interpolating the baseline transmissivity in the band regions. The experimental transmissivity in the bands is then divided by the interpolated transmissivity to correct for the drift. The maximum drift was of the order of 10%, i.e., the transmission spectra obtained had a baseline of between 90% and 110% transmission.

In some cases, there were no optically thin regions of the spectrum outside the bands where the transmissivity was close to unity. This was especially the case for the rotational band of H₂O, which extends to zero on the wavenumber scale. Since the wide-band MCT detector has an useful range of between 600 cm⁻¹ and 4000 cm⁻¹, the transmissivities predicted by HITEMP at these wavenumbers were used to calculate the baseline.

It was also noticed that the locations of the peaks in the experimental transmission spectra did not precisely match those predicted by HITEMP, especially at higher wavenumbers (by a maximum of around 3.5 cm⁻¹). These differences could be due to incorrect FTIR calibration, instrument line function or incorrect line center locations in the HITEMP database. These differences could be of the order of 2 cm⁻¹ [25], although differences of 3.5 cm⁻¹ are somewhat large. Thus, the experimental spectral locations were also modified so as to line up the experimental peaks with those obtained from the HITEMP databank.

An analysis of the experimental error was also performed, as outlined below.

The narrow-band (mean) absorption coefficient $\bar{\kappa}$ is defined as

$$\bar{\tau} = \exp(-\bar{\kappa}xl), \quad (1)$$

where $\bar{\tau}$ is the narrow-band transmissivity, x is the mole fraction of water vapor and l is the path length. The uncertainty in the transmissivity results from uncertainties in temperature, mole fraction and path length. Assuming that the temperature uncertainty affects the narrow-band absorption coefficient only through the gas density,

$$\bar{\kappa} \propto \frac{1}{T}, \quad (2)$$

and

$$\frac{\partial \bar{\kappa}}{\partial T} = -\frac{\bar{\kappa}}{T}. \quad (3)$$

Thus, the root-sum-of-squares (RSS) error (w) in the narrow-band transmissivity may be obtained as,

$$w_{\bar{\tau}} = \sqrt{\left(\frac{\partial \bar{\tau}}{\partial T} w_T\right)^2 + \left(\frac{\partial \bar{\tau}}{\partial x} w_x\right)^2 + \left(\frac{\partial \bar{\tau}}{\partial l} w_l\right)^2}. \quad (4)$$

But

$$\frac{\partial \bar{\tau}}{\partial T} = \frac{\partial \bar{\tau}}{\partial \bar{\kappa}} \frac{\partial \bar{\kappa}}{\partial T} = -\exp(-\bar{\kappa}xl) xl \left(-\frac{\bar{\kappa}}{T}\right), \quad (5)$$

since the temperature does not affect x and l . Thus,

$$w_{\bar{\tau}} = \exp(-\bar{\kappa}xl) (\bar{\kappa}xl) \sqrt{\left(\frac{w_T}{T}\right)^2 + \left(\frac{w_x}{x}\right)^2 + \left(\frac{w_l}{l}\right)^2}, \quad (6)$$

i.e.,

$$\frac{w_{\bar{\tau}}}{\bar{\tau}} = \bar{\kappa}xl \sqrt{\left(\frac{w_T}{T}\right)^2 + \left(\frac{w_x}{x}\right)^2 + \left(\frac{w_l}{l}\right)^2}, \quad (7)$$

or,

$$\frac{w_{\bar{\tau}}}{\bar{\tau}} = -\log \bar{\tau} \sqrt{\left(\frac{w_T}{T}\right)^2 + \left(\frac{w_x}{x}\right)^2 + \left(\frac{w_l}{l}\right)^2}. \quad (8)$$

The uncertainty in temperature was less than 2% at all temperatures. The Micro GC used to measure the H₂O mole fraction was accurate to 5%, while the error in the distance between the bottom of the drop tube to the platinum mirror was around 1 mm, owing to uncertainty in the exact location of the platinum mirror in the furnace, which results from mirror distortion at high temperatures as well as tilting of the mirror during optical alignment. Thus, at the minimum path length of 10 cm, the uncertainty in the length was around 2% (since the path length is twice the minimum 5 cm distance between the bottom of the drop tube and the mirror). Figure 2 shows the resulting uncertainty in the narrow-band transmissivity. As the figure shows, the maximum absolute error in the transmissivity caused by T , x and l is less than 0.03.

4 RESULTS AND DISCUSSION

Figure 3 compares the measurements for pure water vapor ($x_{\text{H}_2\text{O}}=1$) at 1000 K for the rotational band of H₂O with the transmissivities predicted by HITEMP. The figure also shows error bars, which come from the experimental standard deviations of the six different sets of transmission spectra, as described earlier. Though the rotational band essentially goes to 0 cm⁻¹ (or infinity on wavelength scale), the wide-band MCT detector used for these measurements is only accurate above around 600 cm⁻¹, as the larger standard deviations at the low wavenumber end indicate. It is seen from the figure that HITEMP is fairly accurate for this band (above 600 cm⁻¹), though there are regions where the database shows somewhat more absorption than measurement, perhaps indicating the presence of incorrectly extrapolated lines. Data for the rotational band at 600 K were deemed unreliable owing to large inconsistencies between the six transmissivity spectra, which resulted in large error bars and widely varying transmissivities over the band, and thus these data are not presented here.

Figure 4 compares the experimental data for the same band with HITEMP at 1550 K. The database is seen to agree with experiment over most of the band, with minor differences at the high wavenumber end. The error bars are seen to be small at this temperature.

Figure 5 compares the experimental data for pure water vapor at 600 K for the 6.3 μm band of H₂O with the transmissivities predicted by HITEMP. As the figure shows, HITEMP predicts somewhat less absorption than experiment below about 1300 cm⁻¹ and more absorption than experiment above this wavenumber. This trend seems to be reversed at 1000 K (Figure 6), with HITEMP predicting marginally less absorption than experiment beyond around 1500 cm⁻¹. A similar trend is seen at 1550 K, with HITEMP showing more absorption than experiment below about 1300 cm⁻¹, and less absorption above this wavenumber (Figure 7). The standard deviations of the measurement (error bars) are seen to be reasonably small at 1000 K and 1550 K. However, at 600 K, the deviations are much larger, perhaps indicating that these data are less reliable than those at higher temperatures, as the large differences with the HITEMP database in the wings of the 6.3 μm band indicate.

The measured data for the 2.7 μm band of H₂O are compared with HITEMP at the same three temperatures in Figures 8, 9 and 10. At 600 K, HITEMP is seen to show less absorption than experiment in the band wings (below about 3300 cm⁻¹ and above about 4000 cm⁻¹) and more

absorption in the center of the band. The opposite seems to be true at 1000 K (Figure 9), perhaps indicating that the database has incorrectly extrapolated lines, or is missing some high-temperature lines. At 1550 K, agreement between measurement and HITEMP is generally good, with HITEMP predicting somewhat less absorption than the experimental data in the center of the band. This might indicate missing lines in the database. Again, the standard deviations are rather large at 600 K, and the experimental data at this temperature may not be as reliable as desired.

Measured data at 600 K for the 1.8 μm band of H_2O are compared with HITEMP in Figure 11. As the figure shows, HITEMP shows less absorption than measurement over the entire band. The same is true at 1000 K (Figure 12) and at 1550 K (Figure 13), which may indicate missing high-temperature lines in the database.

The figures shown in this section are a representative sample showing the accuracy of HITEMP at elevated temperatures. Many more data for a myriad of conditions may be found in [24].

The measured data for the 2.7 μm band of H_2O were also compared with the data of Phillips [20]. The experimental data at 4 cm^{-1} were degraded to the 25 cm^{-1} resolution of Phillips' data. Figure 14 compares the current measurements with HITEMP and Phillips at 600 K. The error bars of his data were obtained from the standard deviations provided in [20]. The error bar on top was obtained by subtracting one standard deviation from both the k and d' parameters, while the lower one was obtained by adding a standard deviation to the band model parameters. As the figure shows, Phillips' data are in closer agreement with HITEMP over most of the band. However, as stated above, the 600 K data from the current measurements may not be reliable.

Figure 15 compares the degraded experimental data at 1000 K with HITEMP and Phillips. Phillips' data show large error bars, but are in closer agreement with HITEMP. The current measurements show small error bars and predict more absorption than HITEMP, which might indicate missing/incorrectly extrapolated lines in the database.

5 CONCLUSIONS

Narrow-band transmission measurements were made for the rotational, 6.3 μm , 2.7 μm and 1.8 μm bands of H_2O at temperatures ranging from 600 K to 1550 K. These measurements were compared with transmission data obtained from the HITEMP database, as well as with the data of Phillips for the 2.7 μm band of H_2O . Areas of similarity and differences between the experimental and HITEMP data were identified. Phillips' data were seen to be in closer agreement with HITEMP, but showed large error bars at higher temperatures. HITEMP was seen to show slightly less absorption than experiment over most of the bands at all the temperatures considered, indicating that the database may be missing some lines. However, overall data from HITEMP follow experiment rather closely, thus allowing confident use of the database for temperatures up to 1600 K.

ACKNOWLEDGMENTS

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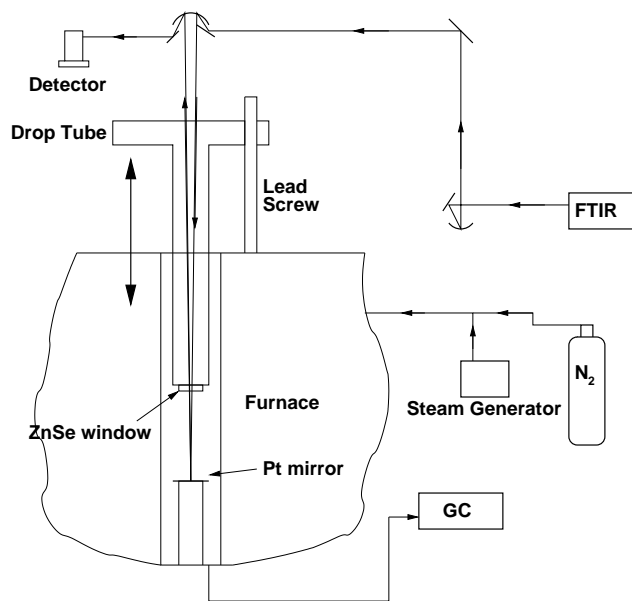


Figure 1: The Experimental Setup (Not to Scale).

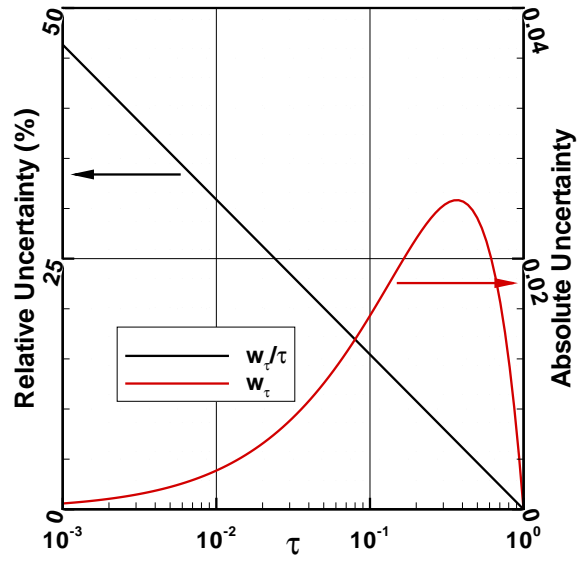


Figure 2: Relative and Absolute Uncertainties in $\bar{\tau}$.

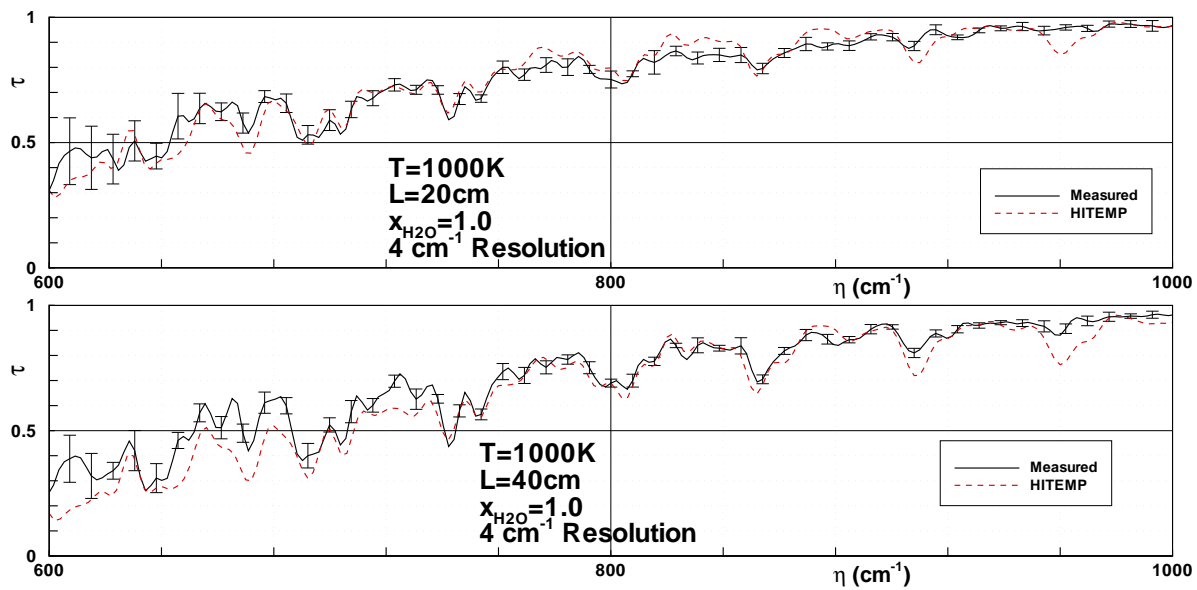


Figure 3: Comparison of experimental data with HITEMP (1000K, rotational band of H₂O).

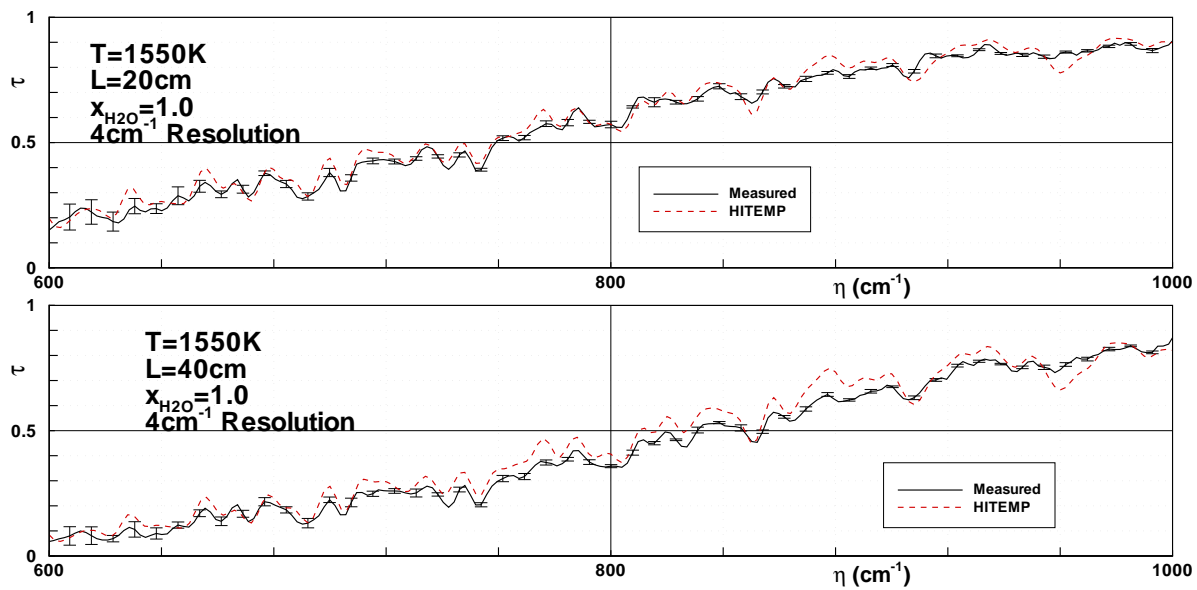


Figure 4: Comparison of experimental data with HITEMP (1550K, rotational band of H_2O).

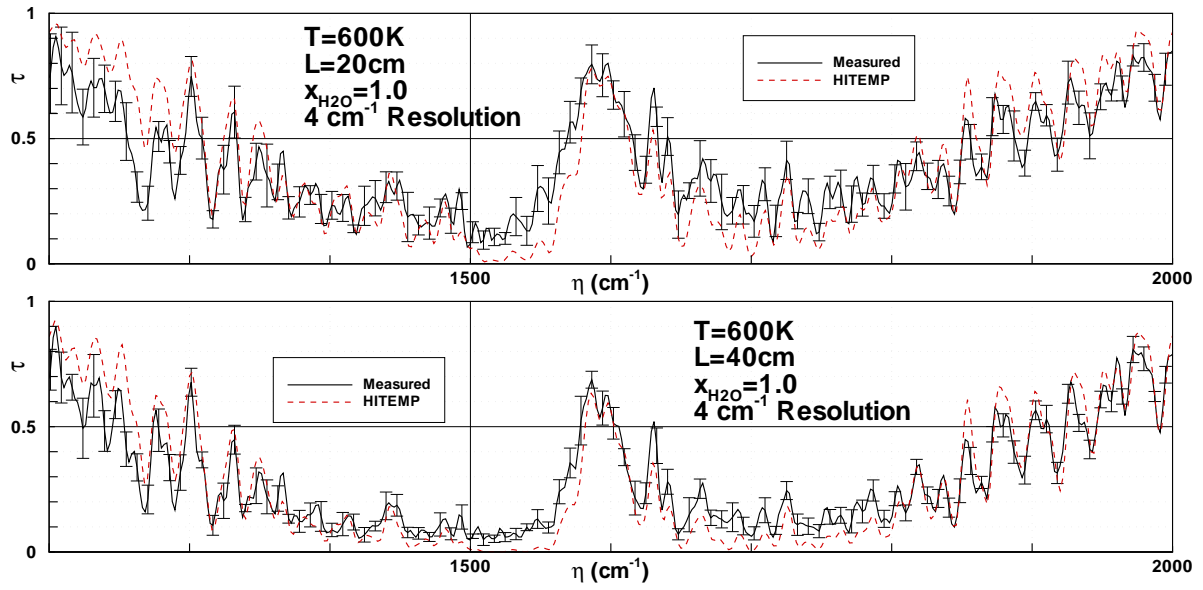


Figure 5: Comparison of experimental data with HITEMP (600K, $6.3\ \mu\text{m}$ band of H_2O).

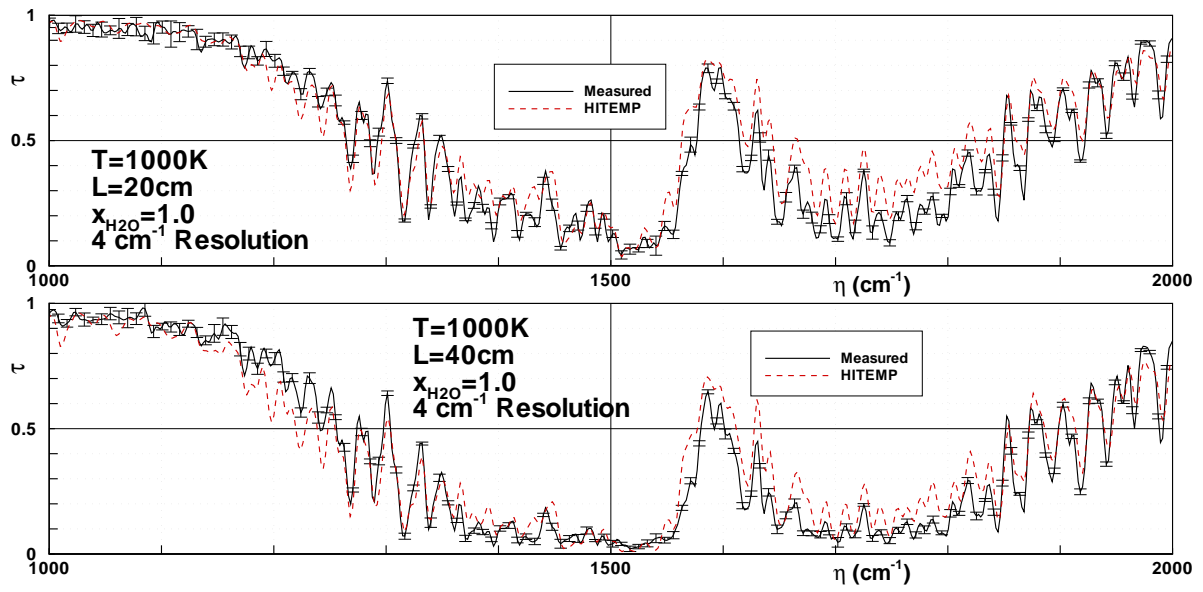


Figure 6: Comparison of experimental data with HITEMP (1000K, 6.3 μm band of H_2O).

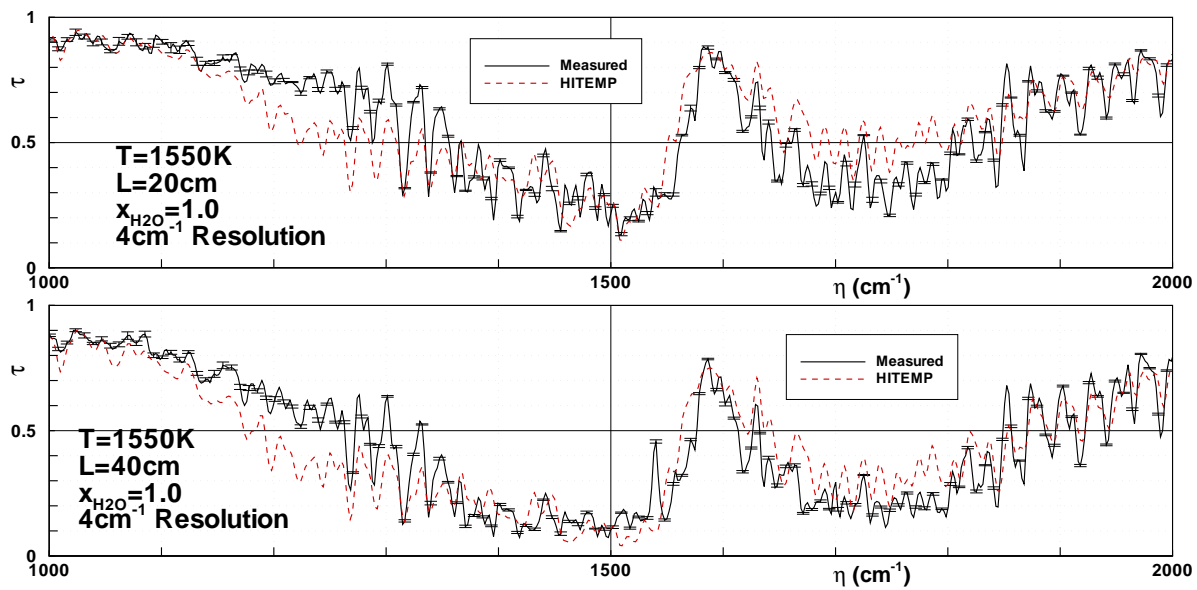


Figure 7: Comparison of experimental data with HITEMP (1550K, 6.3 μm band of H₂O).

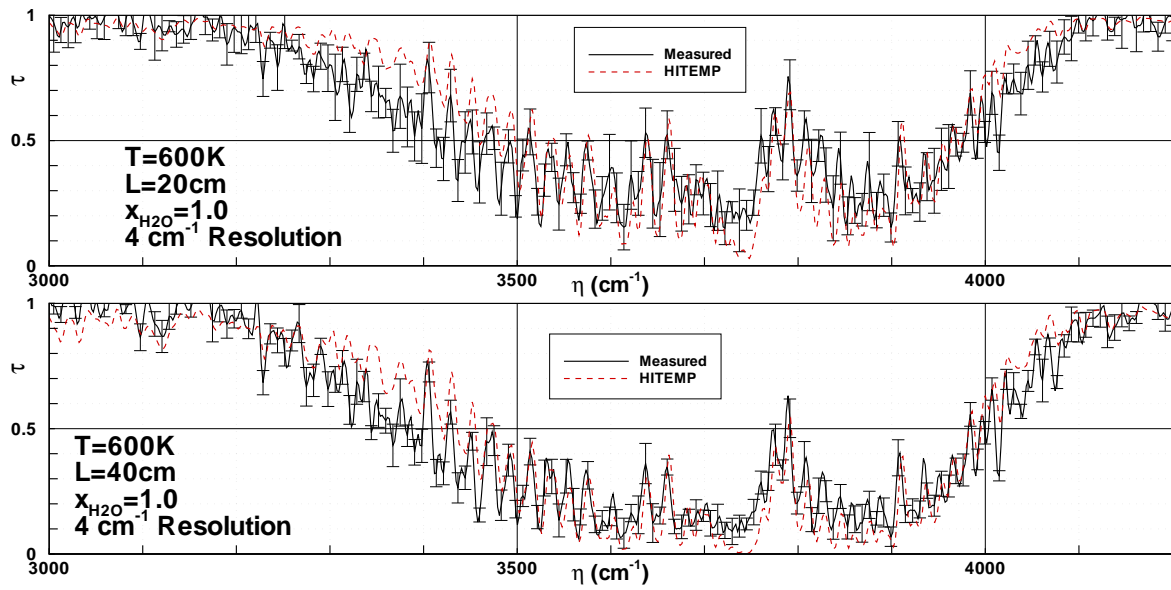


Figure 8: Comparison of experimental data with HITEMP (600K, $2.7 \mu\text{m}$ band of H_2O).

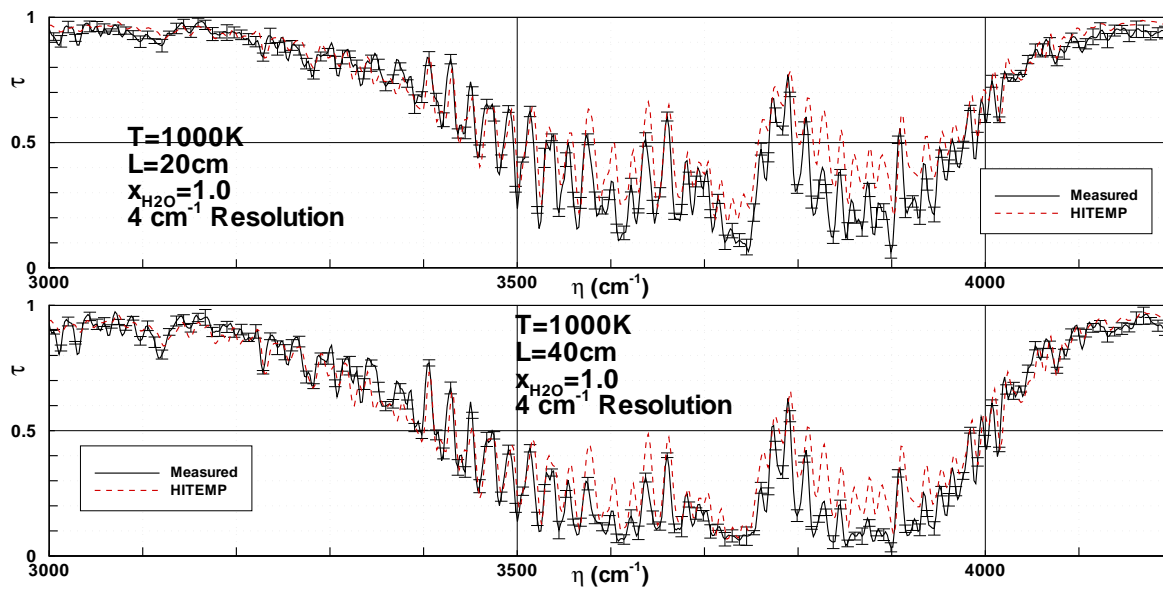


Figure 9: Comparison of experimental data with HITEMP (1000K, 2.7 μm band of H_2O).

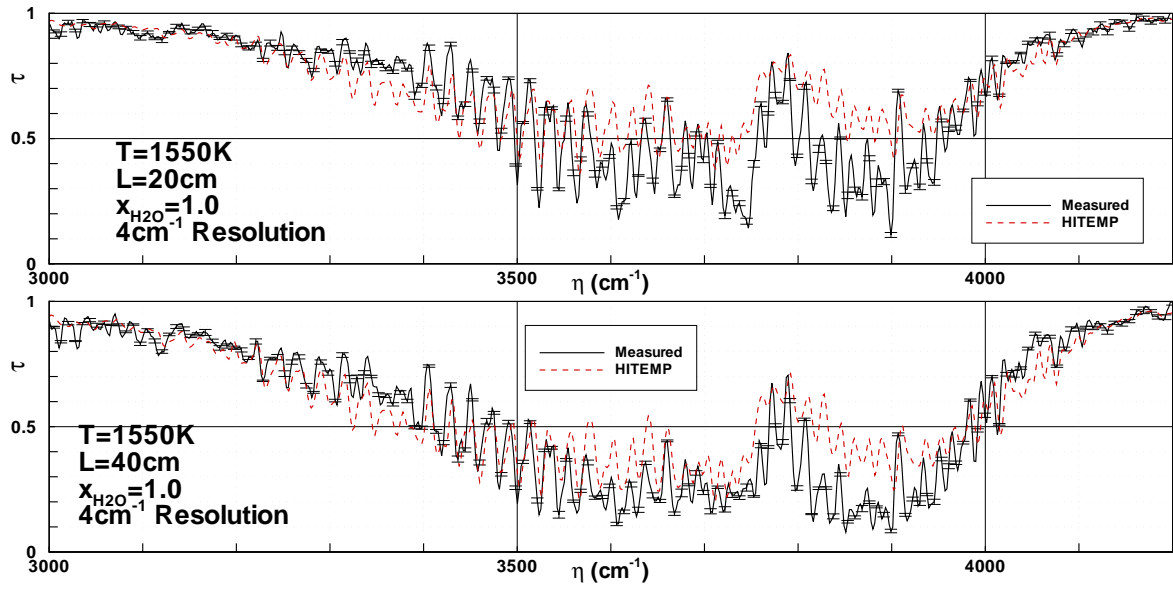


Figure 10: Comparison of experimental data with HITEMP (1550K, 2.7 μm band of H₂O).

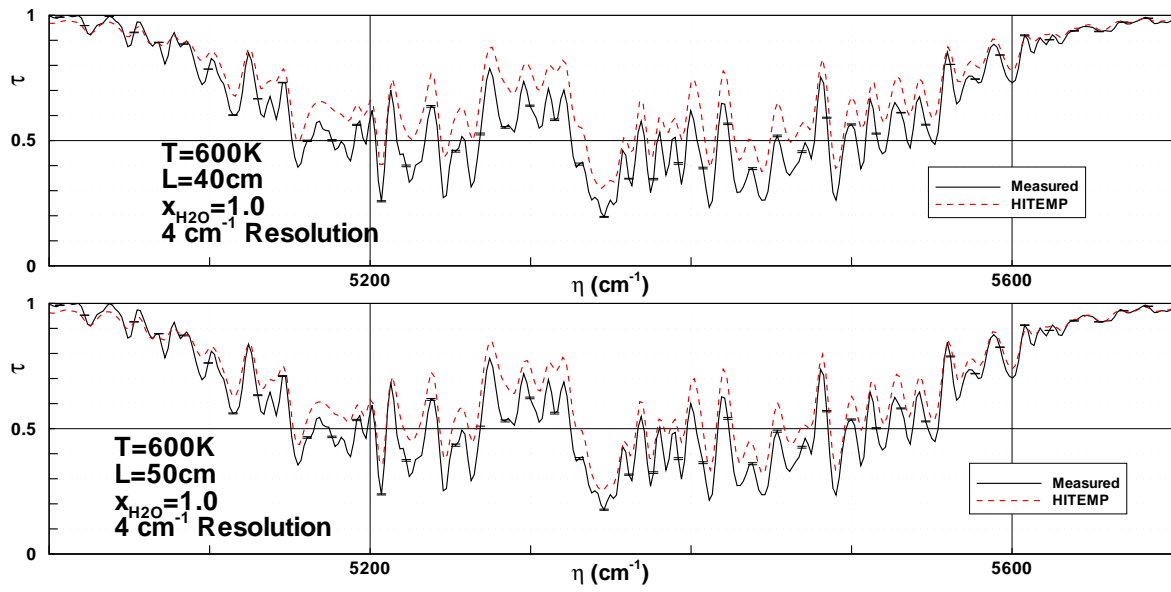


Figure 11: Comparison of experimental data with HITEMP (600K, 1.8 μm band of H_2O).

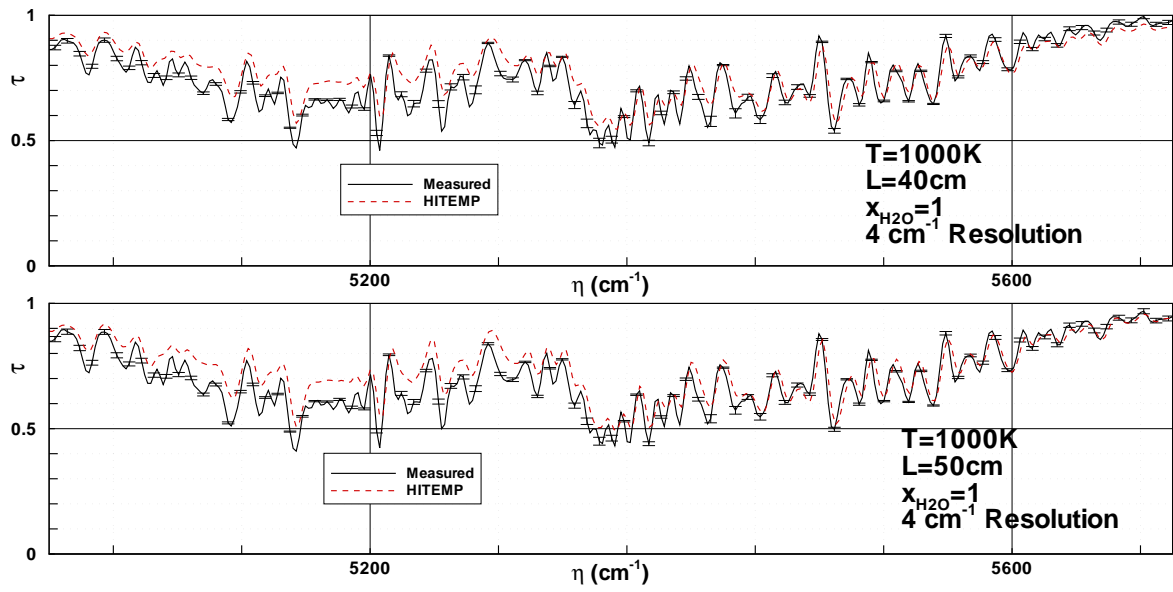


Figure 12: Comparison of experimental data with HITEMP (1000K, 1.8 μm band of H_2O).

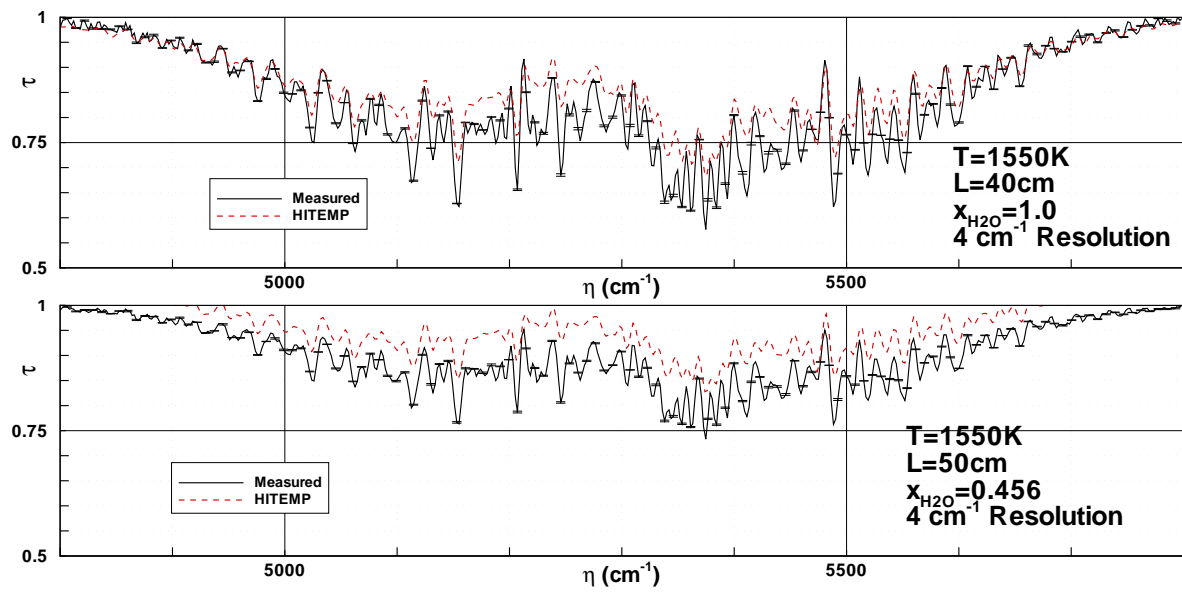


Figure 13: Comparison of experimental data with HITEMP (1550K, 1.8 μm band of H_2O).

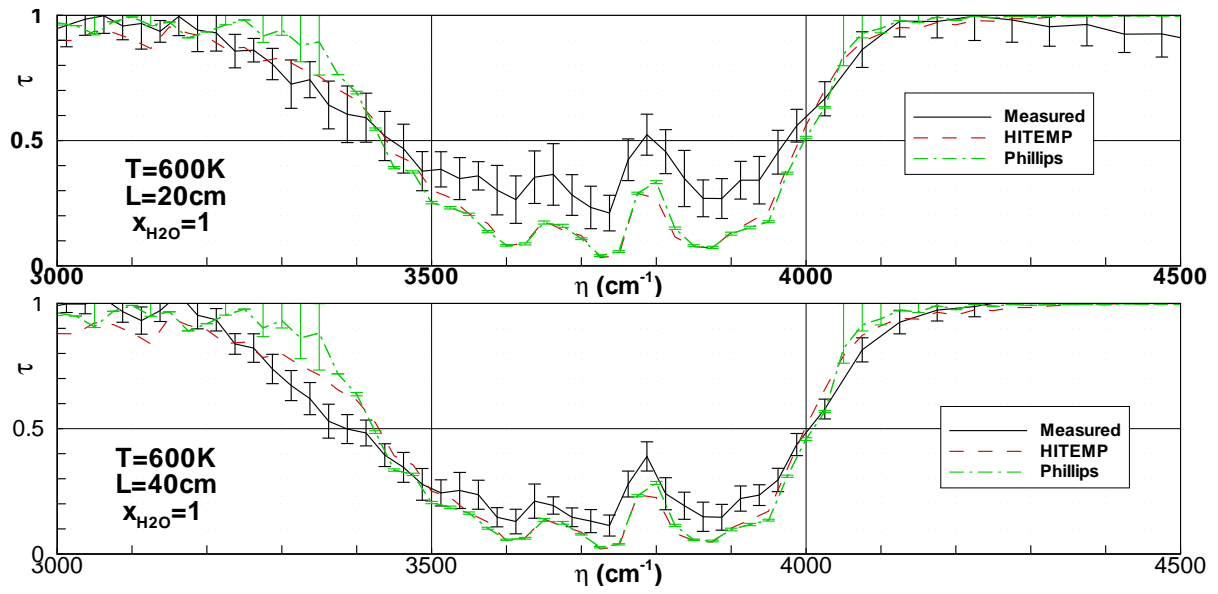


Figure 14: Comparison of experimental data with HITEMP and Phillips (600K, $2.7 \mu\text{m}$ band of H_2O).

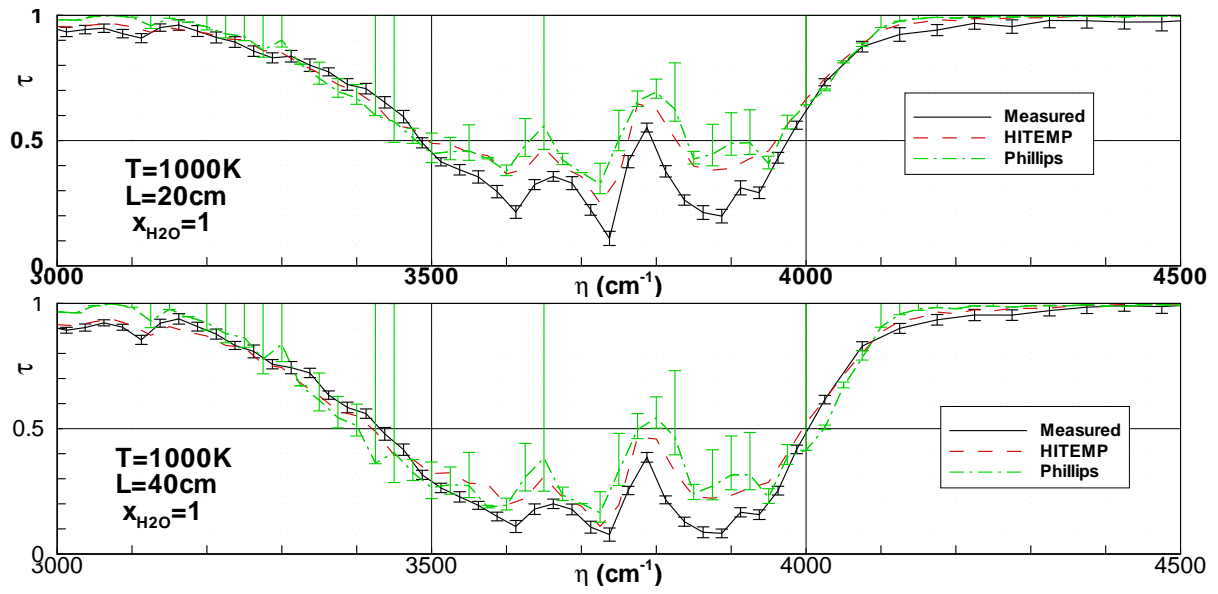


Figure 15: Comparison of experimental data with HITEMP and Phillips (1000K, $2.7 \mu\text{m}$ band of H_2O).