# INFRARED AND MILLIMETER-WAVELENGTH ABSORPTION BY ATMOSPHERIC WATER VAPOR

Attenuation of electromagnetic waves by atmospheric water vapor is an important consideration in a variety of remote-sensing applications. The experimental character of absorption by water vapor in atmospheric window regions at millimeter and 10-, 4-, and 2.2- $\mu$ m wavelengths is surveyed. Also, some of the concepts and models used to characterize this phenomenon are reviewed.

### INTRODUCTION

The propagation of electromagnetic radiation is affected by three major phenomena: absorption, scattering, and turbulence. Absorption by molecular water vapor is the dominant mechanism of tropospheric attenuation at millimeter and infrared wavelengths, especially in the marine environment. Therefore, the understanding and accurate modeling of absorption by this molecule are important to atmospheric remote sensing, infrared imaging systems, long-path laser propagation, electro-optical systems, radar, and atmospheric meteorology. Figure 1a shows the low-resolution infrared transmittance of the atmosphere and demonstrates the importance of water vapor over other atmospheric constituents. The H<sub>2</sub>O absorption bands, along with those of CO<sub>2</sub>, define the atmospheric window regions.

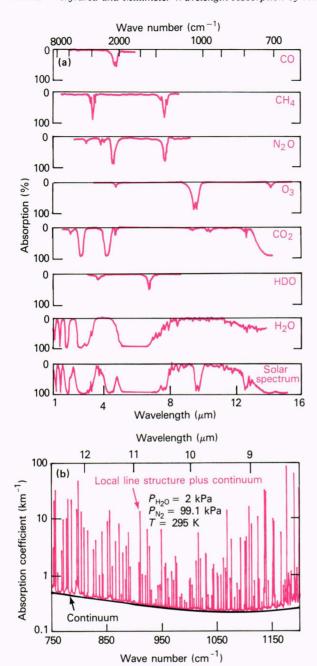
I will concentrate on the water vapor window absorption problem under tropospheric conditions. The main rotational and vibrational bands have been extensively characterized by Benedict and Calfee, <sup>2</sup> Gates et al., <sup>3</sup> and Camy-Peyret and Flaud. <sup>4</sup> The work of those investigators has resulted in a compendium of absorption-line parameters, maintained by the Air Force Geophysics Laboratory, <sup>5,6</sup> which represents a significant contribution to absorption calculations.

The comparatively weak absorption that does occur in the window regions can be described as arising from two distinct sources, local line and continuum absorption, illustrated in the high-resolution spectrum of Fig. 1b for the 10- $\mu$ m window region. Weak absorption bands of CO<sub>2</sub> and HDO (hydrogen deuterium oxide), along with other H<sub>2</sub>O absorption lines in the window regions, compose the local line contribution. The continuum contributes an additional, gradually varying, frequency-dependent background to the total absorption. In 1942, Elsasser<sup>7</sup> recognized in the 13- to 8-μm window region a continuum, which he attributed to the far wings of the strong, nearby rotational and  $v_2$ vibrational-rotational bands of H2O. Further verification of this nonlocal line absorption feature was provided by Yates and Taylor, 8 who studied infrared attenuation along horizontal paths at sea level. Solar spectra studies also indicated continuum absorption in the 13- to 8-µm window. 9-11 The nature of the continuum, judged by

those measurements, was uncertain. It could be due to far wings (far from the band center) of strong absorption bands or to scattering and absorption by particulates.

In an effort to determine the cause of continuum absorption in the 13- to 8- $\mu$ m window, Bignell<sup>12</sup> in 1963 examined solar spectra while monitoring the atmosphere for aerosol concentrations and studying CO<sub>2</sub> far-wing contributions. He concluded that the amount of continuum absorption observed could not be explained by aerosol attenuation or far-wing absorption by CO<sub>2</sub>. An attempt was then made to model the continuum by far wings of the bordering H<sub>2</sub>O bands. The important contribution from this initial work was the realization of major water vapor contributions to the continuum. A second paper by Bignell<sup>13</sup> in 1970 described a careful examination of water vapor absorption in the window regions by use of a multiple-traversal absorption cell and grating spectrometer. Two important characteristics of the 13- to 8-µm window were noted: (a) a large ratio of water vapor self-to-foreign-gas broadening ability (see Eq. 3 below) and (b) a strong negative temperature dependence. Neither of these findings was anticipated on the basis of the far-wing approaches of Bignell's 1963 paper. 13 Also reported by Bignell 13 was a similar, but much weaker, continuum absorption in the 4-μm region.

Since those initial experimental efforts to characterize the water vapor continuum, many measurements have been made. They fall into three categories: (a) measurements within the earth's atmosphere or field measurements, (b) laboratory measurements using a long-path cell and a spectrometer, and (c) laboratory measurements using a long-path cell or a photoacoustic cell and a laser. Although continuum absorption was first observed through long-path field measurements, its precise characterization requires control and knowledge of the propagation path. The effects of turbulence, particulate scattering, temperature variations, and partial-pressure variations simply cannot be precisely determined in a field measurement. Thus, definitive measurements can only be conducted in the laboratory. Spectrometer measurements determine the frequency dependence of the window regions, i.e., local lines and continuum absorption.



**Figure 1**—(a) Low-resolution solar spectrum compared with laboratory spectra of atmospheric gases. (b) Local line structure plus continuum in the 10-μm region.

Laser measurements are limited to discrete frequencies, but because of the laser's higher power and stability, greater accuracy can be obtained; this is particularly true for photoacoustic techniques. Laboratory transmission measurements require very long path lengths (~1 km or longer) and thus are difficult to obtain. The photoacoustic cell, on the other hand, is compact—about 30 cm—but still maintains considerable sensitivity.

As a result of these experiments, a good characterization of the window regions exists today. An excellent review of the field is given by the recent work of Hinderling et al. <sup>14</sup> They emphasize the 8- to  $14-\mu m$  window re-

gion, which, along with the millimeter-wave window, is the most extensively measured. A review of the current experimental data for all the window regions is given in the next section, followed by a brief review of the theoretical and empirical models used to explain the experimental data.

#### EXPERIMENTAL DATA

The experimental data are presented in terms of the absorption coefficient, k, as a function of frequency (or the wave number,  $\nu$ ), temperature, T, water vapor partial pressure,  $P_a$ , and foreign-gas partial pressure,  $P_{N_2}$ . The absorption coefficient is related to the transmittance,  $\tau$ , by

$$\tau = e^{-kL} \,, \tag{1}$$

where L is the path length. A general empirical form for the absorption coefficient used to represent the data <sup>14</sup> is

$$k(\nu) = \frac{P_a}{RT} (C_N P_{N_2} + C_s P_a)$$
 (km<sup>-1</sup>), (2)

where  $C_s$  is the self-broadening coefficient for water vapor (representing the contributions of water vapor to k),  $C_{\rm N}$  is the nitrogen broadening coefficient,  $P_{\rm N_2}$  is the pressure of nitrogen, and R is the ideal-gas constant. The equation can be conveniently rewritten to obtain

$$k(\nu) = \frac{C_{\rm N}}{RT} (P_a) (P_{\rm N_2} + BP_a),$$
 (3)

where  $B = C_s/C_N$  is the dimensionless self-broadening coefficient. Near line center, B has the value of 5. The experimental data to be surveyed are of this form; however, in the real atmosphere, the effects of oxygen broadening must also be included.

# Millimeter-Wave Window

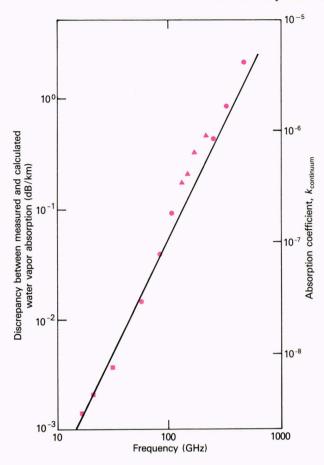
Figure 2<sup>15-19</sup> shows continuum absorption from 10 to 1000 GHz (total absorption minus local lines). The solid line represents an empirical formula given by Gaut and Reifenstein. <sup>16</sup>

$$k_{\text{continuum}} = (1.08 \times 10^{-6}) \rho_a \left(\frac{300}{T}\right)^{2.1} \times \left(\frac{P_T}{101}\right) f^2 \quad (\text{km}^{-1}) ,$$
 (4)

where  $\rho_a$  is the water vapor density (g/m³),  $P_T$  is the total pressure (kPa), and f is the frequency (GHz). The plotted points indicate experimental data. The formula correctly demonstrates the frequency dependence of the continuum but not the temperature and pressure dependence. More recent work by Liebe and Layton²0 uses a continuum formula, fitted to experimental data at 138 GHz, of the form

$$k_{\text{continuum}} = (6.12 \times 10^{-7}) f^2 \left(\frac{300}{T}\right)^{2.4}$$

$$\times P_a \left[P_f + 33.8 \left(\frac{300}{T}\right)^{0.6} P_a\right] \quad (\text{km}^{-1}), (5)$$



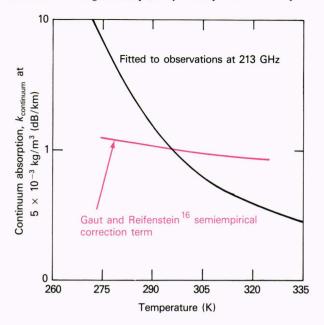
**Figure 2**—Discrepancy between measured and calculated water vapor absorption is indicated by the plotted points, and the empirical correction term given by Eq. 4<sup>16</sup> is shown by the line. The plotted points are ( $\blacktriangle$ ) from Becker and Autler, <sup>17</sup> ( $\spadesuit$ ) from Frenkel and Woods, <sup>18</sup> and ( $\blacksquare$ ) from Burch <sup>19</sup> for T=300 K, P=101 kPa, and  $\rho=10^{-3}$  kg/m³.

where f is in gigahertz, T is in kelvins, and  $P_a$  and  $P_f$  ( $P_f = P_T - P_a$ ) are in kPa. A strong dependence on the water vapor partial pressure is shown (B >> 5). On the basis of the work by Liebe and Layton, <sup>20</sup> the parameter B grows as the frequency decreases from 833 to 110 GHz. Unfortunately, only two temperatures, 282 and 300 K, were used to determine the temperature dependence. A more complete temperature study at 213 GHz by Llewellyn-Jones <sup>21</sup> shows a significantly stronger temperature dependence, illustrated in Fig. 3. <sup>21</sup>

In summary, the millimeter-wave continuum falls off as frequency squared, has an enhanced self-broadening contribution that grows with decreasing frequency, and has a strong negative temperature dependence.

## The 8- to $12-\mu m$ Window

Figure  $4^{21}$  shows the self-broadening water vapor continuum coefficient,  $C_s$ , as a function of frequency between 9 and 14  $\mu$ m at 296 K. The coefficient decreases exponentially as the frequency increases. The same functional dependence is exhibited by  $C_N$ , the nitrogen broadening coefficient, as shown in Fig. 5,<sup>22</sup> but the rate of decrease is more rapid. Thus,  $B (= C_s/C_N)$  in-



**Figure 3—**The temperature dependence of the excess absorption at a water vapor density of  $5 \times 10^{-3}$  kg/m³ is compared with that predicted from the Gaut and Reifenstein 16 semiempirical model.

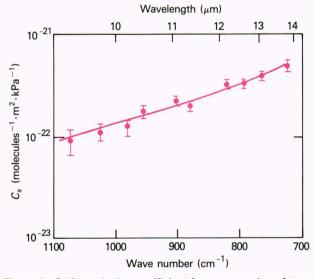


Figure 4—Self-broadening coefficient for wave numbers from 700 to 1100 cm  $^{-1}$  at 296 K.

creases as the frequency increases away from the rotational band. Long-path White cell and photoacoustic laser measurements indicate that B can be quite large. Values ranging from 100 to 600 have been measured in the 10- $\mu$ m region with  $CO_2$  lasers.  $^{14,23,24}$  Figure 6 illustrates a laser measurement at 940.548 cm  $^{-1}$  as a function of the water vapor partial pressure. The total pressure is maintained at 101 kPa with nitrogen.

The observed temperature dependence at  $10.6 \, \mu m$  features a rapid decrease with increasing temperature (Fig. 7), just as in the millimeter window region. The temperature dependence has the functional form

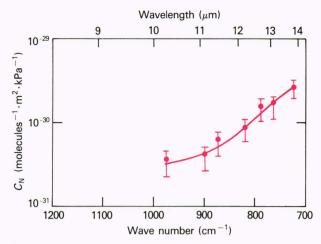
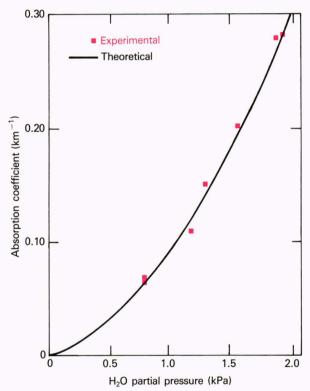


Figure 5— $N_2$  broadening coefficient for wave numbers from 700 to 1200 cm  $^{-1}$  at 296 K.



**Figure 6**—Comparison of experimental and theoretical  $\rm H_2O$  partial-pressure dependence at 940.548 cm $^{-1}$  for T=296 K. The total pressure is 101 kPa;  $\rm N_2$  is used to make up the difference between 101 kPa and the  $\rm H_2O$  partial pressure.

$$C_{\rm s} \propto e^{b/T}$$
. (6)

This result at 10.6  $\mu$ m is also consistent with other measurements in the 10- $\mu$ m region and at 14.3  $\mu$ m. <sup>14,22</sup>

# The 3- to 5- $\mu$ m Window

The 3- to  $5-\mu m$  continuum region has a completely different frequency dependence compared with the millimeter and 8- to  $12-\mu m$  regions. Figure 8b displays an essentially parabolic dependence with a minimum at a

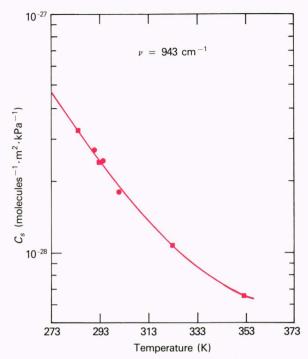
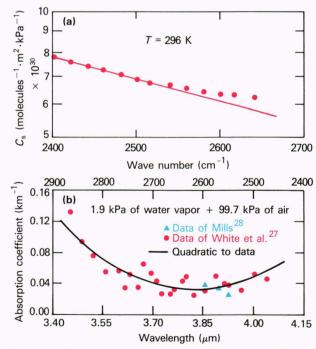


Figure 7—Measured values of the self-broadening coefficient,  $C_s(\nu)$ , as a function of temperature near 10.6  $\mu$ m for pure water vapor samples. The symbol  $\bullet$  indicates the data from Peterson<sup>25</sup> at the 10.6- $\mu$ m White cell measurements. The symbol  $\bullet$  gives the results of Aref'ev and Dianov-Klokov.<sup>26</sup>



**Figure 8—**The 4- $\mu$ m continuum region at T=296 K. (a)  $C_s$  versus wave number<sup>22</sup> and (b) the absorption coefficient versus wavelength.<sup>27,28</sup>

wave number of 2600 cm<sup>-1</sup>. <sup>22,27,28</sup> As shown by the spectrometer measurements of Burch and Alt<sup>22</sup> (Fig. 8a), the self-broadening coefficient has an exponential

falloff at wave numbers up to 2550 cm<sup>-1</sup>, then the falloff rate decreases as the frequency increases. Figure 8b shows long-path White cell DF laser measurements, taken under atmospheric conditions, which indicate continuum absorption levels roughly 50% higher than those indicated by Burch and Alt. The level of absorption in the 4- $\mu$ m region is roughly an order of magnitude less than that in the 10- $\mu$ m region. Long-path CO, DF, and HF laser measurements again indicate large values for *B* in the continuum region, ranging from 20 at 5  $\mu$ m<sup>29</sup> to approximately 50 at 4  $\mu$ m<sup>27</sup> and back down to 10 at 3  $\mu$ m. The sum of the sum of

A strong negative temperature dependence is again observed for the self-broadening coefficient. Figure 9a shows the results of long-path spectrometer measurements by Burch and Alt. The temperature dependence at 2400 cm<sup>-1</sup> exhibits exponential falloff similar to that in the 10- $\mu$ m region. However, the curves at 2500 and 2600 cm<sup>-1</sup> show double trends.

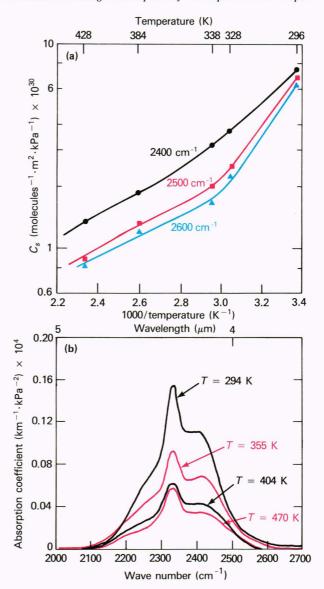
Another interesting feature of this window region is the weak collision-induced absorption band of nitrogen centered at 4.3  $\mu$ m. <sup>31</sup> Figure 9b shows the absorption of nitrogen at 4.3  $\mu$ m over the 296 to 470 K temperature range, as measured at APL. Note that nitrogen absorption also exhibits negative temperature dependence, caused mainly by decreasing density (density  $\approx P_a/RT$ ). Thus, when the nitrogen-broadened water vapor continuum contributions are measured, the absorption band of nitrogen must also be considered.

# The 2.0- to 2.5-µm Window

This window region has not received the same attention as the longer wavelength windows; as a result, no continuum absorption has been previously reported. Recent measurements at APL, however, suggest that continuum absorption does exist. Transmission measurements on hot (T = 685 K) high-pressure (up to 4.8 MPa) water vapor show the continuum absorption in the 2.1and 4- $\mu$ m regions (Fig. 10). <sup>31</sup> Absorption levels at 4  $\mu$ m are consistent with the extrapolated values from the curves in Fig. 9. The point to be made is that a similar continuum absorption process occurs in the 2.1-um region, as shown in Fig. 10. If we assume that an extrapolation to lower temperature is valid, as at 4  $\mu$ m, then a continuum exists in the 2.0- to  $2.5-\mu m$  window that is very similar to the 3- to 5-µm window under normal atmospheric conditions.

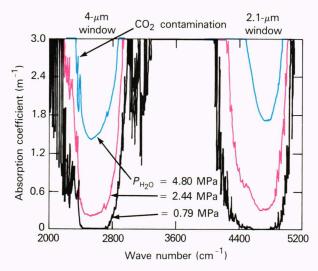
## **CONCEPTS AND MODELS**

No definitive interpretation of continuum absorption by water vapor presently exists. The initial concept of far wings of bordering water vapor bands cannot be verified, because of a lack of line-shape theories valid in the far wing. Clearly, however, far wings must play a role in continuum absorption because of the observed frequency dependence, and work is continuing to find a line-shape theory valid in the far wing. <sup>32-35</sup> The major shortcoming of the line-shape theories is in their failure to predict the observed strong negative temperature dependence characteristic of all the window regions. But this weakness is the very strength of an alternative hy-



**Figure 9**—(a) Plots of the self-broadening coefficients at 2400, 2500, and 2600 cm $^{-1}$  versus the reciprocal of temperature. The symbols  $\blacktriangle$ ,  $\blacksquare$ , and  $\bullet$  represent the experimental data points. (b) The collision-induced absorption band of  $N_2$  at T=294, 355, 404, and 470 K.

pothesis to explain the water vapor continuum: the water dimer. The formation of water vapor dimers has a strong negative temperature dependence that closely matches the temperature dependence of the continuum absorption in the 10- $\mu$ m region.  $^{36,37}$  However, the hypothesis would require a dimer absorption band in every water vapor window, a condition that has not been experimentally found or theoretically shown.  $^{14,38}$  Furthermore, measurements on supersaturated water vapor indicate that dimer absorption is an order of magnitude too small to account for the water vapor continuum at  $10~\mu$ m.  $^{14}$  The dimer model demonstrates the importance of understanding binary water vapor-water vapor interaction to explain the continuum absorption temperature dependence.  $^{14}$ 



**Figure 10**—Pure water vapor absorption coefficient at T=685 K and three different pressures.

## THE APL EMPIRICAL MODEL

Because of the lack of a theoretical foundation for continuum absorption, it is currently characterized by empirical models. One such model, developed at APL and Ohio State University, uses a far-wing model with a semiempirical line shape and the Air Force Geophysics Laboratory's absorption-line data. 39-41 The model has been applied to the 10- and 4-µm window regions and reproduces the experimental data quite well as a function of frequency, pressure, and temperature. An example is the curve in Fig. 6. The water-vapor-broadened far wing falls off less rapidly than the nitrogen-broadened far wing; thus, the self-broadening coefficient grows as the line shape extends from line center. This is the trend observed in the data. The line shape is properly normalized, and the line-center function has been experimentally verified.<sup>42</sup> The far wings (>100 cm<sup>-1</sup> from line center) of a line shape represent a very small percentage of the total area under an absorption line contour. Thus, it is interesting to note that the magnitude of the continuum absorption can be calculated from the far-wing contributions by this model. Contributions to the 10-µm region are dominated by far wings of the very strong rotational band of water vapor on the long-wavelength side of that window, whereas the 4- $\mu$ m continuum absorption arises from the bordering strong vibrational bands of almost equal strength. That difference between sources explains the difference between the frequency dependence of the  $10-\mu m$  continuum region and that of the  $4-\mu m$  continuum region. Unfortunately, the model is not valid in the millimeter region and does not fully represent the recent progress in line-shape theories. The success of this model, therefore, is by no means a statement of the validity of a far-wing interpretation of continuum absorption, but rather a statement of its feasibility.

Another empirical line-shape model with a more satisfactory physical and mathematical character has been applied to  $CO_2$ .<sup>32</sup> The line shape in that model approaches zero frequency as  $f^2$  and has an exponential

far-wing absorption. Thus, the potential to describe the observed frequency dependence in every spectral window of water vapor does exist. The far-wing frequency dependence matches the experimentally observed continuum. Empirical parameters are related to spectral moments and have a physical interpretation. Once the parameters are determined, the model can be tested for physical validity.

Although no definitive interpretation of the continuum exists, the experimental and theoretical evidence indicates that far-wing absorption contributions by the bordering strong water vapor bands play a dominant role. The evidence is based largely on the frequency dependence of the continuum in all four spectral windows reviewed (i.e., the shape of the continuum as a function of frequency and growth of the self-broadening coefficient, B, away from a band as a function of frequency). The shortcoming of the far-wing approach is in predicting the temperature dependence, but the character of a far wing must be driven by close binary interactions much like the creation of a dimer, which does exhibit the observed temperature dependence in the 10- $\mu$ m window region.

## **CONCLUSION**

I have briefly reviewed here the current level of understanding of the water vapor window regions from 10 to 1000 GHz, 8 to 12  $\mu$ m, 3 to 5  $\mu$ m, and 2 to 2.5  $\mu$ m, with emphasis on the continuum absorption observed in each window region. Local absorption-line contributions are important as well, and improved knowledge of position, strength, and half-width must be obtained. In fact, the strong lines of the fundamental rotational and vibrational bands need improved line strength and half-width characterization. The line positions are known quite well.

The observed frequency dependence in each window is consistent with the concept of far wings emanating from the bordering water vapor absorption bands. This connection is considered the most promising basis for modeling continuum absorption. Theoretical line-shape models predict the observed frequency and pressure dependence of absorption, but thus far not the temperature dependence.

Water vapor continuum models are currently empirical. One such model uses a line-shape approach with an empirical far wing and the Air Force Geophysics Laboratory's absorption-line database for the 10- and  $4\mu$ m regions. It represents the observed absorption levels under tropospheric conditions quite well. Therefore, absorption beyond experimentally measured conditions can be predicted with reasonable confidence. Although this model is not conclusive proof of the nature of continuum absorption, it is a useful engineering tool.

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