THE ABSOLUTE TRANSMISSION OF THE ATMOSPHERE

AT MILLIMETRE WAVELENGTHS

by

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This thesis describes new measurements taken from sea level of the atmospheric absorption of solar radiation in the 3-15 cm⁻¹ (0.66-3.3 mm) range. A Michelson interferometer was used, giving a resolution of 0.4 cm⁻¹ (apodized). A hot black body source at 1220°K calibrated the system.

The observed absorption was compared with the predicted monomer absorption (assuming a Gross line shape) and an estimate of the water vapour dimer absorption derived from laboratory measurements could also be included. The observed absorption values were on average twice as much as that predicted by the monomer theory.

At 7.7 cm⁻¹ (1.3 mm) the absorption was correlated with airmass and water amount but there was considerable variability in the measurements caused by anomalous absorption features. These features often observed between 6.5 and 10 cm⁻¹ (1-1.5 mm) were not simply correlated with water amount or airmass.

The excess absorption (above the monomer prediction), measured in the slant path, was greater than observed under controlled conditions in the laboratory, but less than seen in open air horizontal path measurements.

Thick cumulus congestus clouds attenuated the far infrared signal by 3-6 dB but cirrus and 'fair weather' cumulus clouds had very little effect.

The water vapour in the path was determined by measuring the integrated absorption of three water vapour vibration-rotation absorption bands in the 0.9-1.5 μm wavelength region. The bands were calibrated using radiosonde measurements. The water amount in the zenith was found to be well correlated with surface water vapour density, consistent with a scale height of 1.87 ± 0.13 km.
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1.1 The Study of Atmospheric Radiation

Electromagnetic radiation from the sun reaches the surface of the earth at visible wavelengths, selected infrared wavelengths and at wavelengths beyond one millimetre. The intensity of solar radiation peaks in the middle of the visible 'window', whereas the re-emitted terrestrial radiation peaks in the 10 µm window. The opacity of the latter window is an important parameter when considering radiative processes which occur in the atmosphere. General circulation models make use of this parameter over a wide range of temperatures and pressures to determine heating and cooling rates in the atmosphere.

Bignell et al. (1963) measured absorption values in this window through slant paths in the atmosphere and found values in excess of that predicted by collision broadening of nearby absorption lines. The water vapour dimer \((H_2O)_2\) has been suggested by Viktorova and Zhevakin (1967) as a possible cause of this 'anomalous absorption', but recent measurements by Zavody et al. (1979) at ten microns indicate this explanation may be over simplified.

Figure 1.1 shows that beyond the 10 µm window the water vapour in the atmosphere produces high absorptions up to millimetre wavelengths where it begins to transmit radiation again. For the window region from 1 to 1.6 mm the water vapour dimer absorption is predicted to be appreciable (Viktorova and Zhevakin, 1967), so that absorption measurements made in this window region may provide some insight into this anomalous absorption phenomenon. A number of observations of atmospheric absorption in this window have been carried out, both at high altitudes and sea level.
Fig. 1.1  The atmospheric transmission from ultra-violet to centimetre wavelengths for a vertical path to space. Typical amounts for every atmospheric constituent are assumed. The left hand side of this spectrum is based mainly on Goody (1964), the spectral regions bracketed indicate that the transmission was deduced from measurements presented in this thesis.
1.2 Transmission Measurements from Mountain Sites

To investigate if atmospheric absorption could be predicted above a particular location, measurements have been made from a variety of high altitude sites.

Gebbie et al. (1968) measured atmospheric absorption from the Jungfraujoch observatory (elevation 3589 m) in Switzerland. They recorded solar spectra using a Golay detector and Michelson interferometer and found unexplained absorptions between 5 and 10 cm\(^{-1}\) (where cm\(^{-1}\) represents the wavenumber defined as the reciprocal of the wavelength in cm).

Mather et al. (1971) made sky emission measurements from White Mountain, California (elevation 3810 m) using a Fabry-Perot interferometer and cooled bolometer. They bound no appreciable anomalous absorption between 7.5 and 14 cm\(^{-1}\).

Folt et al. (1971) recorded solar spectra from Mauna Kea, Hawaii (elevation 4200 m) using a Michelson interferometer and cooled bolometer. These observations suggested that anomalous absorption was present below 10 cm\(^{-1}\).

Gimmestad et al. (1972) made observations from Baja, California (elevation 2820 m) with a Michelson interferometer and Golay detector. The solar spectra show significant anomalous absorption between 6 and 10 cm\(^{-1}\). The same experimental arrangement was used at Mount Evans, Colorado (elevation 4300 m) to record sky emission spectra (Gimmestad et al. 1977) and the transmission spectrum deduced shows anomalous features below 10 cm\(^{-1}\).

Moffat et al. (1977) made solar measurements with a Michelson interferometer and Golay from Mauna Kea. The water amount in the path was also determined at the same time by recording near infrared spectra.
They found appreciable variability in the transmission at 7.7 cm\(^{-1}\) which was not simply correlated with the water in the path. The zenith angle dependence of the absorption was also complex, although for mountain sites it is questionable whether the water vapour would be horizontally stratified.

Hills et al. (1978) took sky emission measurements from Tenerife (elevation 2400 m) using a polarising interferometer which was absolutely calibrated with a cold black body source. They found that the absorption was correlated with precipitable water measured using a spectral hygrometer and scaled linearly with air mass. The anomalous absorption appeared to be much less at Tenerife.

The above observations indicate that anomalous absorption may vary appreciably from site to site.

1.3 Transmission Measurements from Sea Level

Transmission measurements through slant paths taken from low altitude sites consist mainly of single frequency radiometer measurements with the precipitable water inferred from the surface water vapour density. Ulaby and Straiton (1969) found an attenuation at 8 cm\(^{-1}\) given by 
\[
A(\text{dB}) = 0.6 \rho \quad \text{where} \quad \rho \quad \text{is the surface water vapour density in g/m}^3.
\]
They used a cooled bolometer looking at the sun, with a wire mesh filter arrangement which allows them to scan between 6 and 10 cm\(^{-1}\).

Wrixon and McMillan (1978) using a superheterodyne radiometer system to detect sky emission, found an attenuation of 
\[
A(\text{dB}) = 0.35 \rho \quad \text{at} \quad 7.7 \text{ cm}^\text{-1}, \quad \text{considerably less than the former result.}
\]

Plambeck (1978) presented a large number of measurements taken from an altitude of 1000 m with a radiometer operating at 7.5 cm\(^{-1}\), using both sky emission and solar radiation. He found the attenuations obtained from either method to differ by less than 0.2 dB. His theoretical relation
of \( A(dB) = 0.5 \rho \) looked a reasonable fit to the observations, although a large amount of variability was observed about the mean value.

Johnson et al. (1970) made solar measurements with a radiometer at 7.15 cm\(^{-1}\) and found the relation \( A(dB) = 0.28 \nu \), where \( \nu \) was the water in the path in millimetres precipitable measured with a near infra-red hygrometer.

Absolute transmission spectra from sea level have not been reported but a few spectra have been taken of solar radiation through the atmosphere. Harries and Burroughs (1970) and Harries and Ade (1972) made spectral measurements using a Michelson interferometer and various helium-cooled detectors from Queen Mary College, London. Both series of spectra show anomalous features appearing from 7-10 cm\(^{-1}\), similar to those seen at higher sites.

1.4 Horizontal Path Measurements

A few laboratory and open air transmission measurements have been made which are relevant to slant path measurements.

Frenkel and Woods (1966) made measurements from 3 to 10 cm\(^{-1}\) in a resonant cavity with nitrogen present as a broadening gas to simulate atmospheric conditions. They observed absorptions in excess of that predicted by collision broadened lines. Burch (1968) made similar measurements at higher frequencies and found an anomalous absorption component which increased with frequency, which was confirmed by Bohlander (1979).

Emery et al. (1979) made measurements from 4-15 cm\(^{-1}\) in an open air absorption cell and found on average absorptions which were twice that seen in laboratory measurements, under controlled conditions. This suggests a non-equilibrium phenomenon may be present in the real atmosphere.
1.5 Basic Definitions and Units Used

The measurements described in this thesis were solar spectra recorded from sea level through a slant path for two particular wavelength regions.

Near infrared spectra from 0.9 to 1.8 μm were recorded to measure the water amount in a slant path through the atmosphere; they are described in Chapter 5. The water amount is always quoted in millimetres precipitable which is defined as the depth of liquid water that would be obtained if all the water vapour molecules in a column of unit area were condensed out. Millimetres precipitable are related to other commonly used units as follows:

\[ 1 \text{ mmppt} = 0.1 \text{ g/cm}^2 = 3.35 \times 10^{21} \text{ molecules/cm}^2 = 124.5 \text{ atmo-cm (at S.T.P.)} \]

The length of the slant path through the atmosphere was measured by the airmass which for zenith angles less than 75° is given to within 1.5% by:

\[ a = \sec(Z) \quad \ldots \quad 1.1 \]

where \( Z \) is the angle between the zenith and the sun. For zenith angles larger than 75°, corrections for the curvature of the earth and refraction have to be applied as given by Nagle (1974).

The far infrared spectra from 3-15 cm\(^{-1}\) were recorded with an absolute transmission scale. The absorption units used were in decibels defined by

\[ k(\nu) = 10 \log_{10} \left( \frac{1}{t(\nu)} \right) \text{ dB} \quad \ldots \quad 1.2 \]

where \( t(\nu) \) is the transmission from 0 to 1.

The absorption at a specific frequency was measured by averaging the absorption over one resolution width at that frequency.

The predicted monomer absorption through a gas refers to the value calculated by assuming that no gas molecules in the path are bound together to form larger molecular complexes. This monomer theory is generally
accepted to be the best basis for predicting atmospheric absorption at millimetre wavelengths.

Any additional absorption observed is referred to as 'excess absorption' in the text. Some of this excess absorption may be accounted for, in the atmosphere, by the existence of the water vapour dimer, which is simply two water molecules joined together by a weak bond. The remaining excess absorption is referred to as 'anomalous absorption' in the text since the physical processes causing it are not understood.

1.6 Symbols Used

The symbols commonly used in the text are defined below.

\[ \lambda = \text{wavelength in } \mu\text{m or mm} \]
\[ \nu = \frac{1}{\lambda} = \text{'frequency' in wavenumber } \text{cm}^{-1} \text{ where } \lambda \text{ is in cm} \]
\[ \Delta \nu = \text{spectral resolution in } \text{cm}^{-1} \text{ (defined in Chapter 6)} \]
\[ \kappa(\nu) = \text{absorption coefficient at } \nu \text{ cm}^{-1} \text{ in decibels (dB)} \]
\[ A = \text{absorption at 7.7 cm}^{-1} \text{ in dB} \]
\[ t(\nu) = \text{transmission between 0 and 1 at } \nu \text{ cm}^{-1} \]
\[ I(\nu) = \text{intensity of radiation measured at the earth's surface by a detector looking at the sun} \]
\[ S(\nu) = \text{intensity of solar radiation measured at the earth's surface} \]
\[ E(\nu) = \text{intensity of sky emission radiation measured at the earth's surface} \]
\[ B(\nu) = \text{intensity of black body radiation measured in the laboratory} \]
\[ P(\nu, T) = \text{the Planck function at a frequency } \nu \text{ cm}^{-1} \text{ and temperature } T(0\text{K}) \]
\[ T_s(\nu) = \text{brightness temperature of the sun} \]
\[ T_B = \text{brightness temperature of the black body} \]
\[ T_c = \text{brightness temperature of the chopper blade} \]
\(\sigma(\nu)\) = instrumental response function

\(x = 2X\) = optical path difference between the two paths of radiation in the interferometer \((X = \text{mirror movement})\)

\(P\) = broadening pressure

\(T\) = broadening temperature \((^0 \text{K})\)

\(\rho\) = water vapour density \((\text{g/m}^3)\)

\(P_\text{o}, T_\text{o}\) = standard pressure and temperature \((1 \text{ atm, } 296^0 \text{K in our case})\)

\(n\) = broadening temperature coefficient

\(a\) = airmass

\(\xi\) = zenith angle (degrees)

\(N\) = absorber amount \((\text{molec/cm}^2)\)

\(\omega\) = water in the path in millimetres precipitable \((\text{mmppt})\)

\(\int A_\nu \, d\nu\) = integrated absorption of a band in \(\text{cm}^{-1}\)

\(\nu_{ij}\) = resonant frequency of a molecular transition \((\text{cm}^{-1})\)

\(S_{ij}\) = intensity of the transition per absorbing molecule \((\text{cm}^{-1}/\text{molec/cm}^2)\)

\(\alpha\) = the line half width at half maximum intensity \((\text{cm}^{-1})\)

\(E_i, E_j\) = energies of the lower and upper states \((\text{cm}^{-1})\)

\(J\) = total molecular angular momentum

\(\Omega\) = angular momentum along symmetry axis of molecule

\(\tau = \Omega_A - \Omega_B\) where \(\Omega_A\) relates to a prolate symmetric top and \(\Omega_B\) to an oblate symmetric top

Fundamental constants used:

\(\hbar\) = Planck's constant

\(K\) = Boltzmann's constant

\(c\) = velocity of light
CHAPTER 2 - THEORETICAL PREDICTIONS

2.1 Introduction

The interaction of electromagnetic radiation with individual molecules at infrared wavelengths occurs if the molecule has a dipole moment and is perturbed so that it changes its vibrational or rotational state. If the polar molecule does change its state then radiation is absorbed or emitted at a resonant frequency \( \nu_{ij} \) corresponding to the energy change \( E_j - E_i \) having occurred within the molecule. The Bohr formula:

\[
\nu_{ij} = \frac{E_j - E_i}{h}
\]

where \( h \) is Planck's constant, relates the quantised energy levels within the molecule to the resonant frequencies in the electromagnetic spectrum.

A great deal of theoretical and experimental research has been done concerning the vibrational and rotational energy levels within molecules, so that it is now possible to predict the resonant frequencies and strengths of interactions caused by commonly occurring molecules.

At millimetre wavelengths the absorption of radiation through clear air is dominated by pressure broadened lines from the rotation spectrum of water vapour. Oxygen, the water vapour dimer and to a lesser extent ozone, also absorb radiation at these wavelengths. Scattering by atmospheric aerosols is negligible at these wavelengths. Mie scattering calculations show that scattering by water droplets only becomes significant through clouds or fog. Figure 2.1 shows the predicted transmission spectra for a vertical path through a cloud free atmosphere for typical amounts of each gas. The following sections describe how these theoretical spectra were derived.
Fig. 2.1 Theoretical predictions using the Gross line shape of the transmission through one airmass for the three most absorbing atmospheric gases at millimetre wavelengths. The monomer water amount assumed was 10 mmpppt with a broadening pressure of 0.82 atm. and temperature of 273°K and the dimer amount was an estimated mean value for the atmosphere. The broadening conditions and amounts for the other gases are given in Table 2.1. The spectra do not have an instrumental resolution function included.
The parameters for each line in the frequency range from 1 to 100 cm$^{-1}$ were obtained from McClatchey et al. (1973). The parameters listed are the resonant frequency $v_{i,j}$ (cm$^{-1}$), the intensity per absorbing molecule $S$ (cm$^{-1}$/molec/cm$^2$), the line half width at half maximum $\alpha$ (cm$^{-1}$/atm) and the energy of the lower state $E_i$ (cm$^{-1}$). All these values refer to a temperature of 2960K and a pressure of one atmosphere.

The resonant frequency $v_{i,j}$ is derived from Eqn. (2.1) for each transition and is assumed to be independent of the pressure and temperature values encountered in the atmosphere.

The absolute line intensity $S$ can be calculated from quantum mechanical considerations and can be measured experimentally. It is given by

$$S_{i,j}(T) = \frac{8\pi^3 v_{i,j}^2}{3hc Q_r} \left[ e^{-E_i/KT} - e^{-E_j/KT} \right] \left[ |\mu_{i,j}|^2 \right] \text{ cm}^{-1}\text{molec/cm}^2 \ldots (2.2)$$

where

- $v_{i,j}$ = resonant frequency (cm$^{-1}$)
- $h$ = Planck's constant
- $c$ = speed of light
- $E_i, E_j$ = energies of lower and upper states
- $Q_r$ = the rotational partition function
- $[|\mu_{i,j}|^2] = g' \mu_o |\phi_{i,j}|^2$

where

- $g'$ = nuclear spin factor
- $\mu_o$ = permanent dipole moment of molecule
- $|\phi_{i,j}|^2$ = line strength of transition

The line intensity is only dependent on temperature through the rotational partition function $Q_r$ and the Boltzmann factor in Eqn. (2.2). The partition function for linear and symmetric top molecules can be simplified
to give \( Q^* \propto T \) for linear molecules such as oxygen
\( Q^* \propto T^{3/2} \) for non-linear molecules such as water.

Knowing the intensity \( S \) at a specified temperature \( T^0 \)K the following expression will give the intensity at any other temperature \( T^0 \)K.

\[
S(T) = S(T^0) \left( \frac{T^0}{T} \right)^x \left[ \frac{1 - \exp(-1.4398\nu_i(T)/T)}{1 - \exp(-1.4398\nu_i(T)/T)} \right] \exp \left[ \frac{1.4398 E_i(T - T^0)}{T T^0} \right] \quad \ldots (2.3)
\]

where \( x = 1.0 \) for linear molecules
\( x = 1.5 \) for non-linear molecules.

The line half width at half maximum is a function of both pressure and temperature and is given by

\[
\alpha(P, T) = \alpha(P^o, T^o) \left( \frac{P}{P^o} \right) \left( \frac{T}{T^o} \right)^n \quad \ldots (2.4)
\]

where \( P^o \) and \( T^o \) are the pressure and temperature for which the line parameters are specified. The value of \( n \) varies with rotational quantum number and the geometry of the absorbing molecule. For water vapour line widths Benedict and Kaplan (1959) give a mean value for \( n \) of 0.62, this value was used for every water line in our calculated spectrum. For oxygen the value for \( n \) was taken to be 0.9 from Reber (1972) and for ozone a value of 0.5 was assumed from Liebe and Gimmestad (1978). For air broadened water vapour lines, self broadening becomes noticeable for water vapour pressures above 5 torr. To take this into account, an effective broadening pressure \( (P_e) \) was calculated for water vapour, which could then be used in Eqn.(2.4). It was given by

\[
P_e = P_{air} \left[ 1 + \frac{P_{H2O}}{P_{air}} \left( \frac{\alpha_{H2O}}{\alpha_{air}} - 1 \right) \right] \quad \ldots (2.5)
\]

where
\[
P_{air} = \text{the air broadening pressure}
\[
P_{H2O} = \text{the water vapour pressure}
\[
\frac{\alpha_{H2O}}{\alpha_{air}} = \text{the ratio of self to air broadening} \]
The ratio of self to air broadening, like the temperature coefficient, varies from line to line, a mean value of 4.8 was assumed from Liebe and Gimmestad (1978).

2.3 The Line Shape

The majority of the water vapour and oxygen in the atmosphere exists in the region where pressure broadening determines the line shape. The theoretical problem concerning the shape of pressure broadened lines has been studied by numerous workers such as Ben-Reuven (1969). For atmospheric work the Van Vleck-Weisskopf line shape (Van Vleck and Weisskopf, 1945) and the Gross or kinetic line shape derived by Zhevakin and Naumov (1963) and Gross (1955) are often used, although the validity of these line shapes well away from line centres is open to question. Observations by Burch and Gryvnack (1966) in the near infrared region suggest that the wings of carbon dioxide lines are less absorbing than the Lorentz line shape predicts, and the discrepancy may be different for other frequency regions.

The theoretical studies treat the absorbing molecule as a classical harmonic oscillator which interacts with a 'reservoir' of non-radiating molecules. The Van Vleck-Weisskopf line shape is obtained by assuming a complete interruption of the oscillator's motion, inducing a random phase change over a time period which is much shorter than the resonant period of the line. This gives the following equation for the line shape

\[ F_{VVW}(v) = \frac{1}{\pi} \left( \frac{v}{\nu_{ij}} \right)^2 \left( \frac{\alpha}{(v-\nu_{ij})^2 + \alpha^2} + \frac{\alpha}{(v+\nu_{ij})^2 + \alpha^2} \right) \] ... (2.6)

The Gross line shape makes the assumption that the oscillators do not alter their positions during a collision but their velocities after impact have a Boltzmann distribution. This assumption leads to the Gross line
The Gross line shape appears to give better agreement between observations and prediction (Emery, 1972), (Gibbins, Gordon-Smith and Croom, 1975) than any other line shape based on the impact broadening theory, although large discrepancies still exist away from line centres. Another, more empirical line shape, derived by Schulze and Tolbert (1963) is sometimes used. Figure 2.2 shows typical theoretical spectra derived using different line shape equations. The difference between the Gross and Lorentz line shapes was indistinguishable. The Schulze-Tolbert line shape gave much higher absorptions in the window regions because many high frequency lines were included in the calculation as defined by Table 2.1. When \( v_o \gg v \) the Schulze-Tolbert Line shape gives twice the absorption when compared with the Gross line shape, so that low frequency wings of strong high frequency lines have more effect.

The complete expression for the transmission \( t(v) \) at a frequency \( v \) cm\(^{-1}\) can now be defined as

\[
t(v) = \exp\left[ \frac{8 \pi^3 N^J L_{\xi \nu}}{3 \hbar \omega Q_{\nu}} \left( e^{-E_i/kT} - e^{-E_j/kT} \right) \left( \frac{\mu_{\xi \nu}}{\nu^2} \right)^2 F_G(v) \right] \quad \ldots (2.8)
\]

where \( N \) molecules/cm\(^2\) is a measure of the number of absorbing molecules in the path and the Gross line shape is assumed. To obtain the transmission at a particular frequency, the absorption due to each line at that frequency is calculated and the values are then added together. The criterion for the inclusion of a particular line in the calculation is set out in Table 2.1 and discussed in section 2.7.

2.4 The Instrumental Resolution Function

The calculated spectra do not include the instrumental resolution function. To compare them with the observed spectra the following
Fig. 2.2 Predicted atmospheric transmission spectra derived using different line shape formulations with amounts and broadening conditions as for Fig. 2.1. The solid line represents the Gross line shape and the full Lorentz line shape which are indistinguishable. The dashed spectrum uses the Van Vleck-Weisskopf line shape and the dot-dash spectrum uses the Schulze-Tolbert line shape.
<table>
<thead>
<tr>
<th>Absorbing Gas</th>
<th>Lines included in the calculation</th>
<th>Absorber amount for 1 airmass</th>
<th>Broadening Conditions Assumed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Vapour</td>
<td>All 33 lines from 0-15 cm⁻¹ listed in (1) were used. Lines which have a peak absorption of more than 0.03 dB for 20 mBq/m³ water with a broadening pressure of 1 atm and temperature of 273(K), came into this category. Also 146 lines with frequencies above 15 cm⁻¹ whose low frequency wing contributed more than 4.5×10⁻⁴ dB at 15 cm⁻¹ for the above conditions were included, assuming the Gross line shape (the highest frequency line in this category was at 282 cm⁻¹).</td>
<td>The amount was measured from the near infrared spectra, but the distribution with height was inferred from the radiosonde profile taken at noon. A typical value was 3.36×10⁸ molec/cm³</td>
<td>The broadening pressure, temperature and water vapour density (for self broadening) for each 0.25 km thick layer of the atmosphere below 8 km were derived from the radiosonde profile. From 8 to 100 km values were derived for layers of increasing thickness from the temperate summer version of (4). The density weighted mean values for the whole atmosphere were: P=0.82 atm. T=273(K)</td>
</tr>
<tr>
<td>Oxygen</td>
<td>44 lines were included from 0-15 cm⁻¹ which have a peak absorption of more than 4.3×10⁻⁴ dB for the broadening conditions and amount assumed in the atmosphere. Line data for frequencies below 5 cm⁻¹ came from (1) and above 5 cm⁻¹ from (2).</td>
<td>The mean value was taken to be 4.5×10⁸ molec/cm³ from (3).</td>
<td>Mean values were derived using eq. 2.10 and the temperate summer version of (4). The density weighted mean pressure and temperature assumed were: P=0.50 atm. T=258(K)</td>
</tr>
<tr>
<td>Ozone</td>
<td>All 145 lines listed in (1) from 0-15 cm⁻¹ were included. Lines with peak absorptions of more than 7×10⁻⁴ dB for the broadening conditions and amount assumed in the atmosphere, came into this category.</td>
<td>The mean value assumed was 1.7×10⁸ molec/cm³ derived from (4)</td>
<td>P=0.10 atm. T=230(K) derived as for oxygen.</td>
</tr>
</tbody>
</table>

References
1. McClatchey et al (1973)
2. Gubbin, Burroughs and Bird (1969)
3. Allen (1973)
technique was adopted. A theoretical transmission spectrum was multiplied by an averaged high resolution black body spectrum (with instrumental features) to simulate an observed solar spectrum. This spectrum was then Fourier transformed to produce a simulated interferogram, which was very similar in appearance to the actual solar interferograms measured (figure 3.4). This interferogram was then truncated with an apodising function in exactly the same way as the observations, to produce a theoretical spectrum with the instrumental resolution function included. By adopting this method all artefacts from instrumental features and the Fourier transforming should appear in the theoretical as well as the observed spectra. The apodising function (section 6.1) was chosen so that any appreciable side lobes seen in the theoretical spectrum due to truncating the simulated interferogram were smoothed out.

2.5 The Equilibrium Dimer Absorption

Laboratory measurements described in section 1.4 show absorptions in excess of that predicted by the monomer theory described above. One possible explanation of this absorption first proposed by Viktorova and Zhevakin (1967) was that water dimers were responsible.

The water vapour dimer \((\text{H}_2\text{O})_2\) consists of two water molecules which are loosely bound together by a hydrogen band. The probability of a water molecule occupying a certain region of space is proportional to the water vapour density and the probability of it undergoing a collision with another water molecule is proportional to the density also. This should mean that the number of dimers present in water vapour is proportional to the water vapour density squared. The temperature dependence of the number of dimers should be governed by the energy of formation of the dimer, in which case the number of dimers present may be given by
\[ N_D = \rho^2 e^{-E/KT} \]  

where \( \rho \) = water vapour density \( g/m^3 \)

\( E \) = energy of formation of the dimer (assumed to be 0.12eV Bohlander, 1979)

\( T \) = temperature (°K)

Laboratory measurements of absorption through water vapour under saturated conditions at 300°K (Bohlander* 1979) have produced the absorption contour shown in figure 2.3, which represents the excess absorption observed under controlled conditions. The monomer prediction from which this contour was calculated included the effect of high frequency lines, which can be significant (see section 2.7). Using Eqn. (2.9) the number of dimers present in the atmosphere can be reduced to an equivalent amount under saturated conditions at 300°K. Radiosonde profiles were used to carry out this calculation on each day that measurements were taken. The laboratory contour can then be used to give an estimate of the absorption due to water vapour dimers, in the atmosphere.

2.6 Broadening Conditions in the Atmosphere

2.6.1 Water Vapour

The predicted transmission values in the window regions of the spectrum are strongly dependent on the amount of water vapour in the path, particularly when the transmission has a value around 37% (1/\( \rho \)). In order to obtain an accurate measure of the water amount it was continually monitored during the day using water vapour vibration-rotation absorption bands in the near infrared region (see Chapter 5). By adopting this technique any variability during the day could be taken into account.

To determine the broadening conditions for water vapour the atmosphere was divided up into 250 m thick layers up to 8 km, each having a mean temperature, pressure and water vapour pressure (for the self

---

* The contour finally presented by Bohlander (1979) showed slightly greater absorption, but the conclusions remain the same.
Fig. 2.3 Laboratory measured dimer absorption contour (Bohlander, 1979). The measurements were extrapolated below 7 cm\(^{-1}\), as indicated by the dashed line.
broadening correction). The mean values for each layer were obtained from the radiosonde profile taken 50 km away at Crawley. Above 8 km the atmosphere was divided up into increasingly thicker layers up to 100 km where the water amount becomes negligible. The mean values for these upper layers were taken from the temperate summer version of the U.S. Standard Atmosphere (1966). The water vapour amount measured by the radiosonde in each layer was scaled to give the total water amount observed by the near infrared measurements. The transmission for each layer was then calculated and the individual values were combined to give the transmission in a slant path through the whole atmosphere.

2.6.2 Oxygen

The oxygen amount was assumed to remain at a constant value of $4.5 \times 10^{24}$ molec/cm$^2$ for a vertical path to space from sea level (Allen; 1973). This amount was scaled by the airmass for the particular path observed. The distribution of oxygen with height was assumed to remain constant. The broadening pressure and temperature for a single layer atmosphere were obtained from the temperate summer version of the U.S. Standard Atmosphere (1966). These mean values were derived by using the following relation

$$
\overline{a} = \frac{\int_0^H a(h)\rho(h)\,dh}{\int_0^H \rho(h)\,dh} \quad \text{... (2.10)}
$$

where $a(h)$ is the broadening parameter (i.e. pressure) $\rho(h)$ is the absorber density at height $h$ $H$ is the top of the atmosphere assumed to be 100 km.

The mean broadening parameters derived for oxygen are listed in Table 2.1.
2.6.3 Ozone

The U.S. Standard Atmosphere (1966) was used to determine the ozone amount and mean broadening conditions derived using Eqn.(2.10). The magnitude of ozone absorption in the atmosphere at millimetre wavelengths (see figure 2.1(c)) is small, so that the variability in amount reported by Keating (1978) would not significantly effect the transmission below 10 cm\(^{-1}\).

2.7 The Effect of Various Approximations on the Predicted Absorption

There are a number of approximations which can be made to simplify the theoretical calculations described above, reducing the amount of computer time required.

There were a large number of high frequency water lines above 15 cm\(^{-1}\) included in the calculation because their low frequency wings had a sufficiently high absorption at 15 cm\(^{-1}\) to merit their inclusion. To illustrate how important lines at higher frequencies are to the absorption at 7.7 cm\(^{-1}\) table 2.2 was compiled, which lists the 30 water lines which have the most effect at 7.7 cm\(^{-1}\) in order of decreasing importance, assuming the Gross line shape. Many of the lines are above 30 cm\(^{-1}\) which were not included in the theoretical calculations of Wrixon and McMillan (1978). This explains why their predicted attenuations are significantly lower than the values derived from our calculations.

There is some doubt as to the validity of any line shape far away from line centres. An arbitrary frequency interval could be specified so that any lines with their peaks further away than this frequency would not be included in the calculation. Gaut and Reifenstein (1971) adopt a similar technique by just including the nearest five lines above
### TABLE 2.2

The Effect of Each Water Line at 7.7cm⁻¹ (Gross Line Shape Assumed)

<table>
<thead>
<tr>
<th>Line Frequency (cm⁻¹)</th>
<th>Absorption at 7.7cm⁻¹ (dB)</th>
<th>Percentage of total Absorption</th>
<th>Percentage of Absorption not Included</th>
</tr>
</thead>
<tbody>
<tr>
<td>18.58</td>
<td>.7879</td>
<td>41.788</td>
<td>58.212</td>
</tr>
<tr>
<td>6.11</td>
<td>.1658</td>
<td>8.795</td>
<td>49.417</td>
</tr>
<tr>
<td>25.08</td>
<td>.1221</td>
<td>6.476</td>
<td>42.941</td>
</tr>
<tr>
<td>36.10</td>
<td>.1055</td>
<td>5.598</td>
<td>37.343</td>
</tr>
<tr>
<td>38.29</td>
<td>.0863</td>
<td>4.576</td>
<td>32.767</td>
</tr>
<tr>
<td>12.68</td>
<td>.0773</td>
<td>4.097</td>
<td>28.669</td>
</tr>
<tr>
<td>55.70</td>
<td>.0555</td>
<td>2.935</td>
<td>25.734</td>
</tr>
<tr>
<td>57.26</td>
<td>.0429</td>
<td>2.276</td>
<td>23.459</td>
</tr>
<tr>
<td>37.14</td>
<td>.0328</td>
<td>1.741</td>
<td>21.718</td>
</tr>
<tr>
<td>32.95</td>
<td>.0274</td>
<td>1.453</td>
<td>20.265</td>
</tr>
<tr>
<td>14.94</td>
<td>.0274</td>
<td>1.452</td>
<td>18.813</td>
</tr>
<tr>
<td>47.05</td>
<td>.0271</td>
<td>1.439</td>
<td>17.374</td>
</tr>
<tr>
<td>10.85</td>
<td>.0235</td>
<td>1.247</td>
<td>16.127</td>
</tr>
<tr>
<td>40.28</td>
<td>.0218</td>
<td>1.157</td>
<td>14.970</td>
</tr>
<tr>
<td>55.46</td>
<td>.0212</td>
<td>1.132</td>
<td>13.847</td>
</tr>
<tr>
<td>88.08</td>
<td>.0202</td>
<td>1.072</td>
<td>12.775</td>
</tr>
<tr>
<td>40.99</td>
<td>.0195</td>
<td>1.033</td>
<td>11.742</td>
</tr>
<tr>
<td>92.53</td>
<td>.0155</td>
<td>1.021</td>
<td>10.920</td>
</tr>
<tr>
<td>38.46</td>
<td>.0128</td>
<td>1.068</td>
<td>10.240</td>
</tr>
<tr>
<td>7.53</td>
<td>.0123</td>
<td>1.013</td>
<td>9.585</td>
</tr>
<tr>
<td>72.19</td>
<td>.0113</td>
<td>1.013</td>
<td>8.984</td>
</tr>
<tr>
<td>100.50</td>
<td>.0113</td>
<td>1.013</td>
<td>8.384</td>
</tr>
<tr>
<td>62.30</td>
<td>.0090</td>
<td>1.047</td>
<td>7.909</td>
</tr>
<tr>
<td>75.52</td>
<td>.0089</td>
<td>1.047</td>
<td>7.437</td>
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<tr>
<td>53.44</td>
<td>.0087</td>
<td>1.047</td>
<td>6.974</td>
</tr>
<tr>
<td>79.77</td>
<td>.0078</td>
<td>1.047</td>
<td>6.563</td>
</tr>
<tr>
<td>73.26</td>
<td>.0065</td>
<td>1.047</td>
<td>6.218</td>
</tr>
<tr>
<td>74.11</td>
<td>.0064</td>
<td>1.047</td>
<td>5.880</td>
</tr>
<tr>
<td>64.02</td>
<td>.0057</td>
<td>1.047</td>
<td>5.580</td>
</tr>
<tr>
<td>82.15</td>
<td>.0054</td>
<td>1.047</td>
<td>5.293</td>
</tr>
<tr>
<td>121.90</td>
<td>.0048</td>
<td>1.047</td>
<td>5.039</td>
</tr>
<tr>
<td>127.00</td>
<td>.0046</td>
<td>1.047</td>
<td>4.794</td>
</tr>
<tr>
<td>150.50</td>
<td>.0042</td>
<td>1.047</td>
<td>4.573</td>
</tr>
<tr>
<td>99.03</td>
<td>.0036</td>
<td>1.047</td>
<td>4.381</td>
</tr>
<tr>
<td>104.60</td>
<td>.0036</td>
<td>1.047</td>
<td>4.193</td>
</tr>
<tr>
<td>99.10</td>
<td>.0034</td>
<td>1.047</td>
<td>4.014</td>
</tr>
<tr>
<td>132.70</td>
<td>.0033</td>
<td>1.047</td>
<td>3.839</td>
</tr>
<tr>
<td>68.06</td>
<td>.0030</td>
<td>1.047</td>
<td>3.678</td>
</tr>
<tr>
<td>59.95</td>
<td>.0029</td>
<td>1.047</td>
<td>3.526</td>
</tr>
<tr>
<td>20.70</td>
<td>.0028</td>
<td>1.047</td>
<td>3.376</td>
</tr>
<tr>
<td>111.10</td>
<td>.0028</td>
<td>1.047</td>
<td>3.229</td>
</tr>
<tr>
<td>8.06</td>
<td>.0025</td>
<td>1.047</td>
<td>3.099</td>
</tr>
</tbody>
</table>

Total Absorption at 7.7cm⁻¹ = 1.89dB

(N.B 137 weaker lines not listed were also included in every calculation)
and below the particular frequency of interest. They then add the
effect of high frequency lines and dimer absorption by using an
empirical correction factor valid below 30 cm\(^{-1}\) given by

\[ \Delta \kappa(v) = 4.22 \times 10^{-3} \left( \frac{300}{T} \right)^{2.1} \left( \frac{P}{1000} \right) v^2 \text{dB/km} \quad \ldots \ (2.11) \]

where
- \( \rho \) = water vapour density (g/m\(^3\))
- \( T \) = temperature (°K)
- \( P \) = pressure (mb)
- \( v \) = frequency (cm\(^{-1}\))

Although this method does give reasonable agreement with laboratory
results, it does not help to understand the physical processes causing
anomalous absorption. Since the derivation of Eqn. (2.11) other workers
have carried out calculations using this factor. However, the monomer
calculation to which this correction is added must be as laid out in
Gaut and Reifenstein (1971) for Eqn. (2.11) to be applicable.

As the purpose of this investigation was to measure anomalous
absorption, the high frequency lines were included so there was no possible
doubt as to whether the predicted monomer absorption was sufficient.

Table 2.3 shows the effect which various approximations have on the
absorption at 7.7 cm\(^{-1}\). The inclusion of high frequency lines above
30 cm\(^{-1}\) increases the predicted absorption by 0.6 dB.

Without radiosonde profiles the mean broadening conditions for water
vapour at a certain latitude and time of year can be deduced using Eqn.
(2.10) and an appropriate model atmosphere. The difference in predicted
absorption between the radiosonde and the model atmosphere mean broadening
conditions for a clear day at Slough was 0.26 dB. This difference was
caused by the mean broadening temperature at Slough being 10°C lower than
that assumed in the model atmosphere.
**TABLE 2.3**

The Effect of Various Approximations on the Predicted Absorption

<table>
<thead>
<tr>
<th>Approximations Assumed in the Calculation</th>
<th>Absorption at 7.7 cm⁻¹ (dB)</th>
<th>Difference from Predicted value with no Approximations (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No lines above 30 cm⁻¹ were included. Model atmosphere° mean broadening conditions assumed.</td>
<td>1.19</td>
<td>0.84</td>
</tr>
<tr>
<td>All lines included as defined in table 2.1. Model atmosphere° mean broadening conditions assumed.</td>
<td>1.79</td>
<td>0.24</td>
</tr>
<tr>
<td>All lines included, mean broadening conditions calculated from a typical radiosonde profile using eq. 2.10.</td>
<td>2.05</td>
<td>0.02</td>
</tr>
<tr>
<td>All lines included. Multilayer calculation with the broadening conditions for each layer taken from a typical radiosonde profile. Self broadening was also included.</td>
<td>1.96</td>
<td>0.07</td>
</tr>
<tr>
<td>All lines included, multi-layer theory as above, with ozone absorption included (no approximations)</td>
<td>2.03</td>
<td>0.00</td>
</tr>
</tbody>
</table>

(N.B. Water Amount remains constant throughout)

*U.S. Standard Atmosphere (1966)*

Temperate Summer version used
The comparison between the single layer theory with mean density weighted broadening parameters from radiosonde data and the multi-layer theory with self broadening included was only 0.09 dB. This does suggest that unless great accuracy is required, a good deal of computing time can be saved by only calculating single layer theories with mean broadening parameters.

Finally, the importance of ozone was investigated and was found to make a difference of 0.07 dB to the predicted absorption at 7.7 cm$^{-1}$. Obviously at certain frequencies the effect would be greater than this, as shown in Figure 2.1(c).
CHAPTER 3 - EXPERIMENTAL PROCEDURE

3.1 Comparison of Solar Absorption with Sky Emission Measurements

If a telescope is pointed at the sun, the intensity of radiation measured at the earth's surface is given by

\[ I(\nu) = S(\nu) + E(\nu) \] ...

where

- \( S(\nu) \) is the solar radiation reaching the surface
- \( E(\nu) \) is the radiation emitted by the atmosphere

The earth to space atmospheric attenuation can be deduced by determining \( S(\nu) \) or \( E(\nu) \).

If the intensity of solar radiation reaching the ground \( S(\nu) \) is measured, then the atmospheric transmission is given by

\[ t(\nu) = \frac{S(\nu)}{S_0(\nu)} \] ...

where \( S_0(\nu) \) is the intensity of the sun at the top of the atmosphere.

It can be calculated by assuming the sun to be a black body of known brightness temperature. This method is described in more detail in Chapter 4.

The other method of determining atmospheric transmission is to point the telescope at a blank area of sky measuring \( E(\nu) \) which can be expressed as

\[ E(\nu) = E_B(\nu) t(\nu) + E_A(\nu) \left[ 1 - t(\nu) \right] \] ...

where

- \( E_B(\nu) \) is the cosmic background radiation
- \( E_A(\nu) \) is the intensity of radiation that would be emitted by the atmosphere if it were a black body.

\( E_A(\nu) \) can be calculated by assuming a mean brightness temperature for the atmosphere calculated from radiosonde data using Eqn.(2.10).
The density is replaced by an estimated absorption for each layer. Rearranging Eqn. (3.3) gives the atmospheric transmission as

$$t(v) = \frac{E_A(v) - E(v)}{E_A(v) - E_B(v)}$$

... (3.4)

An uncertainty common to both methods is that a source brightness temperature has to be estimated. The solar brightness temperature has only been measured to an accuracy of about five percent at millimetre wavelengths which, as Hills et al. (1978) point out, introduces an uncertainty of five percent in the transmission value. For high transmissions (0.95) this gives uncertainties of ± 0.05, whereas for lower transmissions (0.40) it becomes ± 0.02 (assuming the Rayleigh-Jeans Law holds).

The uncertainty in the temperature of the emitting molecules does not lead to big uncertainties in the inferred transmission value for high transmissions, but for lower values the temperature distribution with height becomes more important. If an uncertainty of ± 50°C is assumed for the mean brightness temperature of the atmosphere, then at high transmissions (0.95) there is an uncertainty of only ± 0.001, whereas for lower transmissions (0.4) it becomes ± 0.01. To measure high transmissions it appears that sky emission measurements have a much smaller uncertainty, whereas for lower transmissions either method could be used.

The solar method was adopted because the solar signal $I(v)$ was usually an order of magnitude greater in intensity than the sky emission $E(v)$. Another advantage is that the calculation of the observed transmission spectrum is more straightforward with the solar method, as no information is needed about the distribution of water vapour with height. The difficulty with solar measurements is the absolute calibration; it is easier to calibrate a wide angle source, such as the sky, than a discrete source like the sun.
3.2 The Solar Tracking System

There are two possible alternatives for tracking the sun. The first is to mount the telescope, interferometer and detector on an equatorial mount so that the telescope can always be directed towards the sun. The other method is to use a fixed telescope in conjunction with a heliostat system which reflects the solar radiation into a fixed direction throughout the day. The disadvantage found with the first method was that the optics and detector have to be outside (unless one is fortunate enough to possess a dome), which subjects the chopper and interferometer slide to diurnal temperature variations. This can alter the signal recorded and the cut off frequency respectively. Problems may also arise with cooled bolometers because of their marked change of attitude between dawn and dusk. Because of these problems the second method was adopted in one form or another for all of the measurements described, to allow the interferometer, telescope and black body calibration system to be inside the laboratory.

Two forms of heliostat were used, a coelostat on the roof of Imperial College, London, for the early measurements, and a Foucault heliostat for the later observations at Slough. The essential difference between the two systems (Sidgewick 1954) is that with the coelostat the reflected image remains stationary, whereas it rotates about the mirror's axis with the Foucault heliostat. The rotation of the image was no disadvantage for absorption measurements. The Foucault heliostat has the advantage of being able to remain stationary, whereas a coelostat has to be moved with respect to the secondary mirror as the sun's declination changes during the year.

The principle of the Foucault heliostat is shown in Figure 3.1, the length $BD$ must equal $BM$ which allows $BD$ to point towards the sun at all times. The incident radiation is reflected along $BM$ which remains in a constant direction throughout the day. If the
Fig. 3.1 The basic layout of a Foucault heliostat, the North-South direction is in the plane of the page (after Sidewick, 1954).
reflected direction $BM$ is required to be in the meridian (the most symmetrical arrangement) the centre of the mirror $M$ must be south of the polar axis $PB$. If another direction is required, the mirror mount can be rotated about $G$ which is vertically below $B$ until the reflected beam is pointing in the required azimuthal direction. The polar axis $PB$ rotates once every 24 hours to keep $BD$ pointing towards the sun. This form of heliostat is invariably fairly massive to enable the polar axis to be above the mirror.

The optical layout of the experiment, shown in Figure 3.2, was arranged so that the reflected beam was incident on a secondary flat mirror which then reflected it into the laboratory.

A problem common to all heliostats is that for times well away from noon the angle between the meridian and $DM$ in Figure 3.1 is large enough to significantly reduce the area of the heliostat mirror presented to the primary telescope mirror. As the primary mirror must be completely filled with solar radiation for the calibration to remain valid, the system cannot be used well away from noon. This is an important point to be aware of as it not only influences the absolute black body calibration, but also the alternative calibration method obtained from the airmass dependence of absorption.

The flat mirrors in the system act as aperture stops, as illustrated in Figure 3.3. As the sun subtends a finite angle at the earth's surface the effective aperture of the stop (for observations of the complete solar disc) decreases as the distance behind the stop increases. For absolute calibration purposes the telescope mirror should be filled by the 'umbra' region $AA'$ shown in Figure 3.3, as it is only over this area that radiation from every point on the solar disc will be incident upon the mirror. Hence the distance $AB$ should be minimised to meet this criterion and still allow a fairly big diameter telescope mirror to be used. Unless flat mirrors over twice the diameter of the telescope
Fig. 3.2 The optical layout of the system. The moving flat mirror is shown in both configurations.
Fig. 3.3 The effect of the heliostat flat mirror on the reflected solar beam. The further the telescope mirror is away from the heliostat mirror, the smaller the area over which radiation from the whole solar disc can be intercepted.
primary are used and the heliostat-telescope distance is constrained to be less than 10 m, the calibrated observations of the sun are limited to less than four hours before and after noon. A correction factor described in Appendix A was applied to our results well away from noon, to extend the observational period and still allow for this effect.

The mirrors all consisted of glass surfaces coated with a thick layer of aluminium, which was measured to be 35 μm thick by an electron microscope. The skin depth for aluminium at millimetre wavelengths was calculated to be 0.15 μm. Other commonly applied protective coatings were not used because of the uncertainty in their transmission properties in the far infrared region. There was an equal number of mirror surfaces between the detector and either the sun or the black body. Therefore the absolute reflectivity is unimportant as long as the reflectivity of the mirrors outside is the same as the collimating and moving mirrors inside, shown in Figure 3.2. These mirrors had all been re-aluminised prior to the observations. The anomalous absorption observed did not increase with time after the mirrors were aluminised, which suggests that any deterioration of the mirror surfaces outside was insignificant at far infrared wavelengths.

3.3 The Interferometer

The far infrared spectra were obtained from interferograms produced by a Michelson interferometer. To increase the transmission at longer wavelengths a thick mylar beamsplitter was used, which had its first transmission minimum at 15 cm\(^{-1}\). To remove the shorter wavelengths the diverging beam leaving the interferometer was reflected through a right angle by a wire mesh (39 wires/cm) which strongly attenuated all frequencies above 30 cm\(^{-1}\). A variety of filters listed in Table 3.1 were also used with the cooled detector, which ensured that all frequencies above 50 cm\(^{-1}\) remained undetected. The path difference was incremented
<table>
<thead>
<tr>
<th>Location of Filter</th>
<th>Filter Material</th>
<th>Temp. of Filter</th>
<th>Stop Band (Trans. &lt; 1%)</th>
<th>Frequency at which Trans. becomes 50%</th>
</tr>
</thead>
<tbody>
<tr>
<td>In Front of Bolometer Element</td>
<td>Crystal Quartz</td>
<td>2(K)</td>
<td>370-500 cm(^{-1})</td>
<td>323 cm(^{-1})</td>
</tr>
<tr>
<td></td>
<td>Black Polythene</td>
<td>77</td>
<td>above 3300 cm(^{-1})</td>
<td>357</td>
</tr>
<tr>
<td></td>
<td>T12</td>
<td>77</td>
<td>133-625 cm(^{-1})</td>
<td>87</td>
</tr>
<tr>
<td></td>
<td>Calcium Fluoride</td>
<td>77</td>
<td>263-770 cm(^{-1})</td>
<td>222</td>
</tr>
<tr>
<td>In front of Primary Telescope Mirror</td>
<td>Thin Black Polythene Sheet (dustbin liner)</td>
<td>300</td>
<td>above 3300 cm(^{-1})</td>
<td>357</td>
</tr>
<tr>
<td>At Exit of Interferometer</td>
<td>Wire Mesh 39 wires/cm</td>
<td>300</td>
<td>above 30 cm(^{-1})</td>
<td>25</td>
</tr>
</tbody>
</table>
once a second to give a cut-off frequency of 95 cm\(^{-1}\). The screw thread which governed the motion of the moving interferometer mirror was checked for periodic errors. The measured error was found to have a negligible effect on the transmission spectrum, as shown in Appendix B.

The interferometer was placed so that the entrance stop was at the focus of an off-axis reflecting telescope, as shown in Figure 3.2. The preliminary series of measurements at Imperial College were obtained using a Golay pneumatic detector, but the solar signal was only detectable above the noise on very dry days (below 15 mm per day in the path). All of the absolute spectra were recorded using a helium cooled germanium bolometer (Low, 1961) operating at 20\(^\circ\)K.

The incident radiation was amplitude modulated at 34 Hz by a room temperature chopper. This method of modulation has the advantage that the detector only 'sees' radiation from sources above or below room temperature. Radiation from the interferometer and the surrounding laboratory remains undetected. The disadvantage with amplitude modulation is that the mean signal away from zero path difference is non-zero, causing the interferogram to be sensitive to short time scale variations in solar intensity. All interferograms that were recorded during such variations were not analysed. Another disadvantage with amplitude modulation is that half of the incident radiation is prevented from reaching the detector. Phase modulation (Chamberlain, 1971) removes these disadvantages, but unfortunately with cooled detectors all radiation above the temperature of the element (\(\sim 20\)^\circ\)K) will contribute to the signal. Unless the interferometer is cooled, objects such as the beamsplitter will contribute a large proportion of the signal, causing uncertainties in the calibration.
The modulated output signal from the detector was amplified, passed through a phase-sensitive detector, averaged for one second and then recorded on to paper tape for subsequent analysis by computer. A typical solar interferogram is shown in Figure 3.4. They were phase corrected (Vanasse and Sakai, 1967) and Fourier-transformed in the usual manner to produce spectra on a frequency scale.
Fig. 3.4 A typical solar interferogram (solid line) together with the apodising function applied (dashed line).
4.1 The Solar Spectrum

The solar spectra \( I(\nu) \) all contain a component of sky emission radiation, as shown in Eqn. (3.1). The solar radiation component can be written as

\[
S(\nu) = \sigma(\nu) \left[ P(\nu, T_0(\nu)) t(\nu) - P(\nu, T_{chop}) \right] \quad \ldots (4.1)
\]

where
- \( \sigma(\nu) \) = the instrumental response function
- \( P(\nu, T) \) = the Planck function for a temperature \( T^\circ K \)
- \( t(\nu) \) = the atmospheric transmission from 0 to 1
- \( T_0(\nu) \) = the brightness temperature of the sun
- \( T_{chop} \) = the brightness temperature of the chopper.

The brightness temperature of the sun increases with wavelength in the millimetre region (Vernazza et al. 1973), (Stettler et al. 1975), (Gezari et al. 1973) and (Shimabukuro and Stacey, 1968). A best fit line through the observations of solar temperature gave

\[
T_0(\nu) = \left[ 3.51 - \log_{10} (\nu) \right] 2.27 \times 10^3 \ (^\circ K) \quad \ldots (4.2)
\]

where \( \nu \) is between 3 and 12 cm\(^{-1}\).

The sky emission component \( E(\nu) \) reduced the observed intensity, because the emission from the cold sky is of opposite sign to the solar signal, when a room temperature chopper is used.

A sky chopper was tested which eliminated \( E(\nu) \) by oscillating the solar image across the interferometer entrance stop so that solar and sky radiation was compared with just sky radiation, ensuring that only the solar component was detected. This technique was not permanently adopted because the telescope secondary mirror, which oscillated the image, could see around the primary mirror during part of its oscillation, as shown in Figure 4.1. This allowed room temperature radiation to reduce the sky emission signal. Ideally the heliostat mirror should be oscillated to
Fig. 4.1 This illustrates how radiation at room temperature reduces the sky radiation component. This is a disadvantage with this form of sky chopper.
overcome this problem, but this was not feasible because of its size.

The technique finally adopted was to take a solar interferogram to give \( S(v) + E(v) \) and then tilt the heliostat mirror slightly to look at a region of sky close to the sun and record a sky emission interferogram to give \( E(v) \). The emission spectrum could then be subtracted from the solar spectrum and assuming no significant atmospheric changes occurred during the time taken to record both interferograms, the solar component \( S(v) \) should then be given.

4.2 The Black Body Spectrum

To calibrate the interferometer, a hot black body source of known temperature \( T_B \) was used. The image of this source when focussed on to the entrance stop, was circular and of the same diameter as the solar image. The black body spectrum often referred to as a 'background spectrum' is given by

\[
B(v) = \sigma(v) \left( P(v, T_B) - P(v, T_c) \right) \quad \ldots (4.3)
\]

The Rayleigh-Jeans approximation can be assumed at millimetre wavelengths for brightness temperatures above 200\(^0\)K, which simplifies black body intensity calculations by allowing the Planck functions to be proportional to temperature.

Subtracting the sky emission from Eqn.(3.1) and then dividing the result of Eqn.(4.1) by Eqn.(4.3), the atmospheric transmission spectrum is obtained:

\[
t(v) = \frac{S(v)}{B(v)} \cdot \frac{(T_B - T_c)}{(T_\odot(v) - T_c)} \quad \ldots (4.4)
\]

assuming that the solid angles subtended by the solar and black body radiation are identical. The latter assumption was achieved by the method described below.
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\[
B(v) = \sigma(v) \left[ P(v, T_B) - P(v, T_c) \right]
\]  \hspace{1cm} (4.3)

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\[
t(v) = \frac{S(v)}{E(v)} \frac{(T_B - T_c)}{(T_B - T_c) - (T_o(v) - T_c)}
\]  \hspace{1cm} (4.4)

assuming that the solid angles subtended by the solar and black body radiation are identical. The latter assumption was achieved by the method described below.
4.3 The Calibration

A black body source was placed at the focus of a 46 cm diameter collimating mirror with a focal length of 165 cm. The focal length was chosen so that the source subtended the same angle as the sun does at the earth's surface.

At the front of the interferometer in the focal plane of the telescope was placed a circular aperture stop, as shown in Figure 3.2. The diameter of this stop was slightly smaller than the diameters of the solar and black body images which were focussed on to the stop. This ensured that the radiation transmitted through the stop originated from only the sun or black body at visible and near infrared wavelengths. At longer wavelengths (above 1 mm), Fraunhofer diffraction effects from the telescope primary mirror spread out the images, allowing some radiation from the surrounding regions of either source to go through the stop. The solar disc is surrounded by a cold sky, whereas the black body disc has a warm surround so that the calibration would be in error, giving more absorption than was actually present at the longer wavelengths. A theoretical and experimental investigation into diffraction effects which influence the calibration are described in Appendix C. Interferograms were always recorded with the visible solar image or the black body image (which appeared as a dull red disc) centred on the stop.

The calibration of the system was carried out many times during the day, to eliminate any possible doubts concerning changes in the spectral response of the detector and overall gain of the system. The telescope was normally looking at the heliostat but a large flat mirror could be pushed in front of the telescope which allowed it to look at the collimating mirror. This moving mirror was mounted on a slide so that it could be pushed in to a well-defined position every time. This method calibrates the whole telescope as long as the telescope primary was completely filled with either black body or solar radiation.
The background spectra taken during the day were analysed in the following way. To determine whether any consistent change had occurred during the day, each individual background spectrum was divided by the last background spectrum taken that day. Any changes during the day showed up in the ratio as being significantly greater or less than one. If no significant changes had occurred, the individual spectra were then averaged together to produce a mean background spectrum for that day, which reduced the overall noise level of the background spectrum. The individual background spectra were then divided by the averaged background spectrum in exactly the same way as the solar spectra were. An error analysis for these ratios was calculated in the same way as the errors in the solar spectrum were deduced (see Appendix D). The resultant ratios with error bars were plotted, as shown in Figure 4.2, to be certain that changes from unity were not significantly greater than the error bars calculated.

4.4 The Black Body Source

The black body source was a modified version of the one described by Saiedy and Goody (1959). It consisted of a ceramic spherical cavity 12.5 cm inner diameter and wall thickness of 1.3 cm, mounted on fire-bricks in a flameproof box (Figure 4.3). The sphere was thermally insulated with filter cell powder to ensure a uniform heat loss in all directions. It was heated by a coiled wire embedded in the wall of the sphere. The normal operating temperature was 1220 degrees K which was continually monitored by a thermocouple probe in the centre of the sphere.

The theoretical analysis of black body radiation in a spherical cavity at millimetre wavelengths was discussed by Baltes et al. (1970). When the cavity radius approaches the wavelength, resonances develop which cause deviations from the Planckian radiation distribution assumed in Eqn. (4.3). According to the calculations of Baltes et al. (1970), the
Fig. 4.2 Ratio of an individual black body spectrum over the daily average black body spectrum. The error bars indicated were derived using Eqn. (D.4).
Fig. 4.3 The black body source used (after Saiedy and Goody, 1959) with a photograph of the source, taken looking into the exit pipe.
deviations from black body radiation for our source would be about one percent for a resolution of 0.3 cm$^{-1}$.

The sphere had an exit hole 1.5 cm in diameter, allowing the radiation to escape (introducing this loss factor causes the resonances to broaden). The ceramic cone shown in Figure 4.3 helped to reduce the radiative temperature of the exit hole surround which causes uncertainties in the calibration at longer wavelengths (see Appendix C).

A comparison of the thermal black body source with an enclosed mercury lamp source described by Emery and Gebbie (1977) was made using a Golay detector. There appeared to be no significant spectral difference between the two sources above 7 cm$^{-1}$ (the signal to noise was very poor below this). The radiative temperature of the mercury lamp at millimetre wavelengths was calculated to be 38000K ± 2000K by comparing it with the black body radiation of known temperature.

The black body source was assumed to have an emissivity of one at millimetre wavelengths. It should be noted that if the emissivity had been less than one, any anomalous absorption observed would be increased, since $T_B$ should be reduced in Eqn.(4.4).
CHAPTER 5 - THE MEASUREMENT OF PRECIPITABLE WATER

5.1 Introduction

The amount of water vapour in the atmosphere is an important variable when predicting the transmission of the atmosphere at millimetre wavelengths. There are two methods available for measuring precipitable water through a slant path.

A common but rather expensive method is to use a radiosonde which monitors atmospheric pressure, temperature and humidity as it rises. The total vertical water amount can be calculated from this data and then scaled to the required airmass. The value derived by this method refers to the particular trajectory the balloon followed which is a function of wind direction and strength.

Recently spectral hygrometers have been developed to give an instantaneous water amount in a slant path through the atmosphere, using the sun or moon as a radiation source. Their disadvantage is that the sky must be relatively clear to make measurements since cloud strongly attenuates the solar radiation at the near infrared wavelengths used.

Most spectral hygrometers compare the solar intensity at a wavelength on one edge of a water vapour absorption band with the intensity at an adjacent wavelength where there is no significant water vapour absorption. Greve (1978), Hirst et al. (1970) and Sivertsen and Solheim (1975) describe various forms of this type of hygrometer, which are portable and easy to use.

The measurements of precipitable water described here were obtained using a grating spectrometer. This enabled the areas of complete absorption bands to be measured so that the calibrations obtained would be of use to anyone who could scan across one of these bands. The measured spectra could also be compared with spectra derived using the AFCRL line data (McClatchey et al. 1973). This comparison is one way in
which the bands can be calibrated to give water amount.

5.2 Instrumentation

Solar radiation was focussed on to the entrance slit of the grating monochromator by a small Newtonian telescope and was modulated at 20 Hz with a chopper, as shown in Figure 5.1. The instrument was mounted on an equatorial sun tracker. A gallium arsenide filter blocked all wavelengths below 0.9 μm to allow spectra to be scanned between 0.9 and 1.8 μm. The position of the grating was incremented by a stepper motor once a second. The radiation was detected by a Golay cell. The signal was then amplified by a phase sensitive detector and recorded for subsequent analysis by a computer. Two minutes were required to scan an absorption band, the complete spectrum was recorded in ten minutes.

To remove instrumental features in the solar spectrum, a tungsten filament lamp was focussed on to the entrance slit to give a background spectrum $B(\lambda)$. The lamp was assumed to be a black body source at 2900°K, which was determined by comparing its intensity with a cooler black body source of known temperature. The brightness temperature of the sun was assumed to be 6000°K at these wavelengths (Allen, 1973), and independent of wavelength between 0.9 and 1.6 μm. The Rayleigh-Jeans approximation is not valid at near infrared wavelengths, but the radiation from the sky and room temperature chopper becomes negligible so that equation (4.4) becomes

$$t(\lambda) = \frac{S(\lambda)}{B(\lambda)} \cdot \frac{P(\lambda,T_B)}{P(\lambda,T_G)} \quad \ldots (5.1)$$

The transmission spectra did not have an absolute transmission scale as no attempt was made to match the solid angles from the sun and the tungsten lamp. A typical atmospheric transmission spectrum is shown in Figure 5.2(a). The spectral resolution, governed by the slit width of the spectrometer, was 0.003 μm.
Fig. 5.1 Layout of the near infrared spectrometer.
5.3 The Spectra

Approximately 340 slant path transmission spectra were recorded on 58 days, some from the Appleton Laboratory, Slough and some from the roof of Imperial College in London. They were recorded during cloud-free conditions between November 1976 and June 1978 with airmasses between 1.1 and 7.0. If the far infrared interferograms were being recorded, the near infrared spectra were normally recorded simultaneously.

A typical transmission spectrum is shown in Figure 5.2(a), the three water vapour absorption bands can be clearly seen at 0.94 μm, 1.14 μm and 1.38 μm.

The 0.94 μm band is produced by six transitions which occur from the ground state to the 220, 121, 300, 201, 102 and 003 vibrational levels of the water vapour molecule. The band centres for each transition are 10284 cm⁻¹, 10329 cm⁻¹, 10600 cm⁻¹, 10613 cm⁻¹, 10869 cm⁻¹ and 11032 cm⁻¹ respectively. The 201 band is the strongest with the 121 and 003 bands also contributing significantly.

The 1.14 μm band is due to three transitions which occur from the ground state to the 012, 111 and 201 levels centred at 9000 cm⁻¹, 8807 cm⁻¹ and 8762 cm⁻¹ respectively. The 111 band is the strongest.

The 1.38 μm band is produced by five transitions from the ground state to the 120, 021, 200, 101 and 002 levels centred at 6775 cm⁻¹, 6871 cm⁻¹, 7201 cm⁻¹, 7250 cm⁻¹ and 7445 cm⁻¹ respectively with the 101 band being the strongest.

A weaker oxygen band at 1.27 μm corresponding to the (0-0) transition and a few carbon dioxide bands can also be seen in Figure 5.2(a).

To measure the amount of water in the path the integrated absorption \( \int A_v \, dv \) of a water vapour band is used. It is defined as
Fig. 5.2  A typical near infrared spectrum recorded (A) and a spectrum calculated from the AFCRL tabulations (B) with the appropriate absorber amounts (10 mm ppt of water) and instrumental resolution function ($\Delta \lambda = 0.003 \mu m$). The limits between which the integrated absorption was measured are indicated by the dashed lines. Regions of the spectrum with significant absorption not due to water vapour are marked. The error bars in (A) are an estimate of the random noise on the spectrum.
\[ \int_{v_1}^{v_2} A_\nu \, d\nu = (1 - \overline{T}) \Delta \nu \quad \text{cm}^{-1} \quad \ldots \quad (5.2) \]

where \( v_1 \) and \( v_2 \) are the band limits in cm\(^{-1}\)
\[ \overline{T} = \text{the mean transmission between } v_1 \text{ and } v_2 \]
\[ \Delta \nu = v_2 - v_1 \quad \text{cm}^{-1} \]

The mean transmission for each band was derived by fitting a straight line across the top of the band as shown in Figure 5.2(a). The same procedure was followed with the theoretical spectra described below, which removes any effects due to scattering providing the extinction varies linearly across the absorption bands. Also effects due to the cumulative absorption from the wings of strong water lines at longer wavelengths are removed by this method. Assuming this fitted line represents unit transmission for our purposes, the observed mean transmission \( \overline{T} \) can be calculated between the chosen band limits.

The observations show an increase in scattering at shorter wavelengths. Deirmendjian (1964) derived two haze models with different particle size distributions which can be used in Mie scattering calculations. The calculated extinction coefficient at 0.7 \( \mu \text{m} \) is more than double that at 1.6 \( \mu \text{m} \) for a haze model with a mean particle size of 5 \( \mu \text{m} \), which agreed with the observed scattering increase.

5.4 The Calibration

Three possible ways of relating integrated absorption to precipitable water have been considered.

5.4.1 Laboratory Spectra

The first possible method uses laboratory spectra recorded with known amounts of water vapour in the path at various broadening pressures and temperatures. The relationship between integrated absorption and water
amount for various broadening conditions can then be deduced for each absorption band. Howard, Burch and Williams (1956), (HBW), made measurements in this spectral region but their water vapour amounts were lower than the values commonly occurring in slant paths, for the 0.94 \( \mu m \) and 1.14 \( \mu m \) bands. The 1.38 \( \mu m \) band was measured with higher water amounts, but in slant path spectra an oxygen band at 1.27 \( \mu m \) and a carbon dioxide band at 1.43 \( \mu m \) are within the band limits over which the integrated absorption was measured. As these gases were not included in the laboratory-measured integrated absorption values they will be in error when applied to the slant path spectra, and so a calibration was not attempted using the HBW formulae.

The integrated absorptions measured by HBW were obtained by assuming that the reference spectrum taken with no water vapour in the path represented unit transmission. The difference between the water vapour and the reference spectra then gave the integrated absorption which would have included any scattering and far wing absorption. This method would give agreement with the values derived by fitting a straight line between the band limits if the transmission at the band limits was unity for all water amounts.

5.4.2 Spectra derived from AFCRL Tabulations

The second method involves the calculation of transmission spectra using the line parameters from the AFCRL listing (McClatchey et al., 1973). The spectra were derived according to the usual formulae set out in Chapter 2. The different line shapes gave indistinguishable results at these wavelengths (the Gross line shape was actually used). Water vapour, oxygen and carbon dioxide lines were included in the calculation, all other atmospheric constituents were assumed to have a negligible absorption between 0.9 \( \mu m \) and
**TABLE 5.1**

Sources of Data used to Compute the Near Infra Red Spectra.

<table>
<thead>
<tr>
<th>Absorbing Gas</th>
<th>Lines from (1) included in the calculation between 5350 and 11100 cm⁻¹</th>
<th>Absorber amount for 1 airmass</th>
<th>Broadening Conditions Assumed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Vapour</td>
<td>13,193 lines with a peak absorption of more than 0.03 dB for broadening conditions assumed to the right. No lines were included below 5000 cm⁻¹</td>
<td>As required 5-25mm ppt was observed for a vertical path.</td>
<td>Single layer values were derived from (2) using eq. (2.10). The density weighted mean pressure and temperature were P=0.82 atm. T=277(K)</td>
</tr>
<tr>
<td>Oxygen</td>
<td>152 lines with a peak absorption of more than 4.3×10⁻⁴ dB.</td>
<td>The mean value was assumed to be 4.5×10⁻⁴ molec/cm² from (3)</td>
<td>P=0.50 atm. T=258(K) derived as for water</td>
</tr>
<tr>
<td></td>
<td>1695 lines with a peak absorption of more than 6.3×10⁻⁴ dB</td>
<td>The mean value was assumed to be 7×10⁻⁴ molec/cm² from (3)</td>
<td>P=0.50 atm. T=258(K) derived as for water</td>
</tr>
</tbody>
</table>

References
(3) Allen (1973)
1.6 μm. Table 5.1 sets out the criterion adopted for the inclusion of lines and the amount and broadening conditions for each gas.

A number of spectra were calculated with different broadening conditions to determine if the predicted integrated absorption was sensitive to them. The broadening pressure did alter the integrated absorption but the temperature had very little effect, which means that the calibration obtained would be valid for summer or winter. This is not surprising since the bands are produced by transitions from the ground state. The number of molecules populating this state are only weakly dependent on temperature. This small temperature dependence was also inherent in HBW's results since their empirical relationships do not include temperature as a variable influencing the integrated absorption.

Single layer theories were derived using broadening parameters calculated by Eqn.(2.10). A multilayer calculation was also carried out using an eight layer atmosphere, but as in the far infrared, the transmission differed from the single layer theory by only one percent.

The predicted integrated absorption values calculated for various water amounts can be used to calibrate each band. The values were derived in exactly the same way as for the observed spectra by fitting a straight line over the band. The resolution function of the spectrometer was convolved with the calculated spectrum to give the transmission spectrum shown in Figure 5.2(b). The band limits over which the integrated absorption was measured for each band are also shown and listed in Table 5.2.

5.4.3 Radiosonde

The method commonly adopted to calibrate spectral hygrometers (Robson and Rowan Robinson, 1979) is to obtain simultaneous measurements
TABLE 5.2

Analytical Expressions for Calibration Curves

\[ \log_{10}(w) = A + B \int v \, dv \] (w in mmppt, \( \int v \, dv \) in cm\(^{-1} \))

<table>
<thead>
<tr>
<th>Water Band</th>
<th>Band Limits</th>
<th>A</th>
<th>B</th>
<th>Correction Factor Applied to APCRL Curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \mu m )</td>
<td>( \mu m )</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.94</td>
<td>10640</td>
<td>0.20</td>
<td>0.0053</td>
<td>0.95</td>
</tr>
<tr>
<td>1.14</td>
<td>8772</td>
<td>0.12</td>
<td>0.0043</td>
<td>1.15</td>
</tr>
<tr>
<td>1.38</td>
<td>7250</td>
<td>-0.82</td>
<td>0.0030</td>
<td>1.50</td>
</tr>
</tbody>
</table>

For values of \( w \) between 5 and 70 mmppt
of water amount from a radiosonde. Two ascents a day are made at Crawley, a site 50 km away from Slough.

The water amount was calculated from the aerological data by dividing the atmosphere up into 250 m thick layers and applying the formula given by Solot (1939) to each layer

\[
\bar{\rho} = \frac{216.7 \ e(h)}{T(h)} \text{ g/m}^3
\]

where

- \( \bar{\rho} \) = the mean water vapour density for the layer
- \( T(h) \) = the mean temperature (°K)
- \( e(h) \) = the mean water vapour pressure (mb)

Above 8 km an exponential water vapour distribution was assumed with a scale height of 2 km. The mean densities were multiplied by the thickness of each layer to give the total vertical water amount which was then scaled to the airmass for which the integrated absorptions were measured.

Possible doubts about a systematic difference in water amount above Crawley and Slough were investigated by comparing surface water vapour densities at Heathrow (near Slough) and Gatwick (near Crawley). Heathrow was found to be on average five percent drier than Gatwick. The significance of this difference, however, depends upon the calibration of the wet and dry bulb thermometers and their local environment. The differences above the boundary layer should be less so as the difference was small it was assumed that on average the water amount was the same above both sites.

Figure 5.3(c) shows the measured integrated absorption values for the 1.38 \( \mu \text{m} \) band plotted against water amount from the radiosonde, together with a calibration curve derived from the AFCRL tabulations. This predicted calibration curve for the 1.38 \( \mu \text{m} \) band gave significantly lower water amounts than the radiosonde indicated were present.
Fig. 5.3 Calibration curves for the three bands, relating integrated absorption to precipitable water measured by radiosonde. The dashed line is the prediction from the AFCRL tabulation, which was then scaled by a certain amount to give agreement with the observations (solid line).
This discrepancy has also been evident in horizontal path measurements (Emery et al., 1979) but it was smaller, being only 10%, whereas for the slant path it was 50%. The water in the path was an order of magnitude less in the horizontal path, suggesting that the discrepancy increases with water amount. To bring the predicted calibration curve into agreement with the radiosonde values requires a reduction of 20% in the broadening pressure.

The calibration curves for the other two bands shown in Figure 5.3 (a) and (b) were in closer agreement with the radiosonde although differences still existed. The discrepancy for the 0.94 μm band was in the opposite sense compared to the other bands, which suggests that the broadening conditions were not in error.

Each predicted calibration curve was scaled by a correction factor to give agreement with the radiosonde water amounts. These corrected curves were used to calibrate the bands. The correction factors, together with an analytical expression for the corrected curves, are listed in Table 5.2.

Finally a check on the calibration was made by comparing the 6.1 cm⁻¹ water line at millimetre wavelengths (from which a value for the water in the path can be inferred) with the near infrared water amount obtained at the same time. The values obtained were in reasonable agreement, as shown in Figure 6.4, but uncertainties caused by anomalous absorption prevented an accurate check from being made.

5.5 Measurements of Precipitable Water in the Atmosphere

The variation of precipitable water in the zenith over the eighteen months of observation is shown in Figure 5.4. Considerably less water (7 mmppt) was present on clear winter days than on clear summer days (20 mmppt).
Fig. 5.4 The variation of precipitable water in the zenith on clear days between Nov. 1976 and June 1978. Jan. 77, Mar. 77, Apr. 77, Aug. 77 and Dec. 77 all had less than five measurements in their mean value. No values were recorded in Dec. 76 and Feb. 78.
Fig. 5.5 The variation of precipitable water with airmass. The cross represents the noon radiosonde value and the error bars represent the uncertainty of an individual measurement.
Daily variations in water amount were investigated by plotting the water in the path against airmass. If the atmosphere was horizontally stratified and the water amount was constant, the water should be linearly related to airmass. Fig. 5.5(a) shows a plot recorded one clear afternoon when the water amount was constant. Figure 5.5(b) shows a similar plot with points taken during the morning and afternoon where a change in water amount had occurred. The water amount was often higher in the afternoon because of evaporation from the surface during the day. These changes illustrate that on certain days errors may result when using the radiosonde data taken at noon to obtain the water amount in a slant path.

Another method which has been used to obtain precipitable water values in the zenith at any time, is to measure the surface water vapour density and then assume a scale height of 2 km for water vapour to calculate the total water amount. Reber and Swope (1972) analysed radiosonde data from various sites and came to the conclusion that reliable estimates of precipitable water could not be obtained from surface water vapour density measurements.

Measurements made by Madden (1974) and Robson and Rowan-Robinson (1979) both indicate a good correlation between the two quantities consistent with a scale height of 2 km. Figure 5.6 shows a plot of vertical water amount measured from the integrated absorption of the 1.14 μm band against surface water vapour density. A good correlation existed between both quantities at Slough, although the variability observed would sometimes lead to large errors in the inferred water amount. The best fit line through the points gave a scale height of $1.87 \pm 0.03$ km for water vapour. This correlation may not apply to mountain sites (Moffat, 1978) where the water vapour distribution may be far from horizontally stratified and may not decrease exponentially with height close to the surface.
Fig. 5.6 Vertical water amount measured with the 1.14 μm band, plotted against surface water vapour density. A scale height of 2 km would correspond to the dashed line.
CHAPTER 6 - THE ATMOSPHERIC ABSORPTION MEASUREMENTS

6.1 Introduction

A preliminary series of solar spectra were recorded using a Golay detector from the roof of Imperial College, London. An averaged spectrum for one particularly dry day is shown in Figure 6.7(a).

All of the absolute absorption spectra described below were taken on nine reasonably cloud free days between March and June 1978, from the Appleton Laboratory, Slough, U.K. (elevation 20 m). The values for the vertical water vapour amount varied between 5 and 16 millimetres precipitable, the airmasses from 1.1 to 4.0 and the surface temperatures from 12°C to 30°C. The conditions for each day are listed in Table 6.1. The interferograms were only recorded when the attenuation due to cloud was negligible.

Observations were limited to zenith angles less than 75° during the summer, because the heliostat could not make accurate absolute measurements for larger hour angles (Section 3.2). Effects due to the curvature of the earth and refraction influencing the airmass were less than 1.5% for these zenith angles (Nagle, 1974) and so they were neglected.

Each interferogram took about 10 minutes to record, giving a resolution between 0.5 and 0.38 cm⁻¹. The resolution $\Delta \nu$ (cm⁻¹) is defined throughout as the reciprocal of the optical path difference at which the interferogram was truncated. The apodizing function applied to both observed and theoretical spectra was:

$$f(x) = \cos \left( \frac{\pi x}{2x_{\text{max}}} \right) \quad \phi < x < x_{\text{max}}$$

$$= \phi \quad \text{elsewhere} \quad \cdots \ (6.1)$$

where $x$ is the optical path difference

$x_{\text{max}}$ is the path difference at which the interferogram is truncated.
<table>
<thead>
<tr>
<th>Date</th>
<th>Duration of Observations</th>
<th>Mean Absorption at 7.7 cm⁻¹ (dB)</th>
<th>Mean Water in the Path (mm ppt)</th>
<th>Mean Airmass</th>
<th>Surface Temp at noon</th>
<th>Surface water vapour density</th>
<th>Condition of sky</th>
</tr>
</thead>
<tbody>
<tr>
<td>7/3/78</td>
<td>1310-1625</td>
<td>10 ± 2</td>
<td>38 ± 4</td>
<td>2.95</td>
<td>9°C</td>
<td>5.6 g/m³</td>
<td>Thin Ci</td>
</tr>
<tr>
<td>6/4/78</td>
<td>1015-1655</td>
<td>3.9 ± 2</td>
<td>10 ± 1</td>
<td>1.95</td>
<td>10</td>
<td>5.0</td>
<td>Clear</td>
</tr>
<tr>
<td>7/4/78</td>
<td>0855-1645</td>
<td>4.1 ± 2</td>
<td>11 ± 1</td>
<td>1.85</td>
<td>9</td>
<td>4.8</td>
<td>Thin Ci</td>
</tr>
<tr>
<td>9/5/78</td>
<td>1120-1645</td>
<td>4.9 ± 2.5</td>
<td>20 ± 2</td>
<td>1.64</td>
<td>17</td>
<td>9.4</td>
<td>Hazy, clearer later Clear</td>
</tr>
<tr>
<td>10/5/78</td>
<td>0705-1500</td>
<td>4.0 ± 2</td>
<td>17 ± 1.7</td>
<td>1.45</td>
<td>16</td>
<td>6.4</td>
<td>Clear</td>
</tr>
<tr>
<td>16/5/78</td>
<td>0950-1520</td>
<td>5.3 ± 2.5</td>
<td>16 ± 1.6</td>
<td>1.31</td>
<td>15</td>
<td>7.1</td>
<td>Cu around</td>
</tr>
<tr>
<td>17/5/78</td>
<td>1040-1615</td>
<td>5.5 ± 3</td>
<td>17 ± 1.7</td>
<td>1.40</td>
<td>14</td>
<td>8.3</td>
<td>Hazy</td>
</tr>
<tr>
<td>25/5/78</td>
<td>0910-1640</td>
<td>4.0 ± 2</td>
<td>14 ± 1.4</td>
<td>1.37</td>
<td>17</td>
<td>5.2</td>
<td>Clear</td>
</tr>
<tr>
<td>26/5/78</td>
<td>0900-1355</td>
<td>6.3 ± 3</td>
<td>20 ± 2</td>
<td>1.30</td>
<td>17</td>
<td>7.1</td>
<td>Clear</td>
</tr>
</tbody>
</table>
6.2 The Zenith Angle Dependence of the Absorption

The variation of absorption with airmass can be a check on the black body calibration described in Chapter 4. If the atmosphere is uniform and horizontally stratified the absorption should be linearly related to the airmass. The best fit line through the observed absorption values can be extrapolated to zero airmass. If the black body calibration was correct, the line should give zero absorption at zero airmass.

In practice the atmosphere is not uniform and on some days the water amount in the zenith varies appreciably during the day, as shown by the near infrared measurements (Figure 5.5). For short periods of time (~ few hours) the atmosphere was often more stable, allowing absorption measurements taken late in the afternoon, when the airmass was changing rapidly, to be linearly related to airmass.

Figure 6.1 shows absorption values averaged between 7 - 9 cm\(^{-1}\) plotted against airmass for two days when the water amount remained reasonably constant as measured by the near infrared bands. Both plots show that when the airmass was changing rapidly the absorption values were linearly related to airmass. If the best fit line through the points taken late in the afternoon over a time period of 1½ hours was extrapolated, it was close to the origin on both days. If absorption values taken during the morning were also included, however, the best fit line did not go through the origin because the amount of water in the atmosphere had changed during the day.

This technique can be applied at all frequencies to give a transmission spectrum which is independent of the black body spectrum, as the following analysis shows. The transmission \( t(\nu) \) is related to the absorption coefficient \( k(\nu) \) by the relation

\[
 t(\nu) = e^{-k(\nu)a} \quad \ldots (6.2)
\]
Fig. 6.1 The airmass dependence of the total absorption averaged between 7 and 9 cm\(^{-1}\), for two days when the water amount remained constant. The best fit line through the points taken during the afternoon is also shown.
Substituting 6.2 in 4.3 and taking the natural logarithm gives

$$\ln \left[ \sigma(v) \mathcal{P}(v, T_0(v)) \right] - \ln S_1(v) = k(v) a_1 \quad \ldots (6.3)$$

for an airmass $a_1$ and solar spectrum $S_1(v)$.

To eliminate the first term, another solar spectrum $S_2(v)$ at a different airmass $a_2$ can be recorded and the difference between the two is now independent of $\sigma(v) \mathcal{P}(v, T_0(v))$

$$\ln S_2(v) - \ln S_1(v) = k'(v) (a_1 - a_2) \quad \ldots (6.4)$$

which by rearrangement becomes

$$k'(v) = \frac{\ln S_2(v) - \ln S_1(v)}{a_1 - a_2} \quad \ldots (6.5)$$

where $k'(v)$ is the absorption corresponding to an airmass of $(a_1 - a_2)$. To increase the signal to noise, the natural logarithm of a number of low and high airmass spectra was calculated and then all low airmass spectra were averaged together to give a mean spectrum $\ln S_2(v)$ and similarly with the high airmass spectra. The resultant absorption coefficient was converted back into transmission units using Eqn.(6.2). Figure 6.2 shows a transmission spectrum derived in this way together with an appropriate theoretical spectrum derived in exactly the same manner by averaging individual predicted spectra for each observation. The observed transmission spectrum illustrates that the anomalous absorption increased at higher airmasses, suggesting that the absorber was horizontally stratified.
Fig. 6.2 Measured (solid) and predicted (dashed) zenith angle spectrum for 6/4/78. The ratios of mean airmass and water amounts are indicated (note that the vertical water amount changed during the day). The error bars represent the uncertainty in transmission the resolution was 0.5 cm$^{-1}$. 

$\bar{w} = 9.8/9.4$ mmpp$^t$ 

$\sec(z) = 2.6/1.5$
6.3 The Transmission Spectra

Eighty four individual spectra were recorded from Slough and the spectra for each day were averaged together to give daily averages of transmission with the random noise minimised. However, if any discrete absorptions which only existed for an hour or two were present, they would be smoothed out by this averaging process.

Figure 6.3 shows three typical daily averages for different water amounts. Both water vapour monomer lines at 6.1 cm⁻¹ \((J_T = 2 \rightarrow 3)\) and at 10.9 cm⁻¹ \((J_T = 40 \rightarrow 54)\) and the oxygen line at 3.96 cm⁻¹ \((\Omega = 1 \rightarrow 1, J = 0 \rightarrow 1)\) are clearly seen and in good agreement with the monomer theory.

The monomer theory with the equilibrium dimer absorption contour added (Section 2.5) was also plotted. This gave better agreement with observation although there was still some anomalous absorption remaining, especially for high water amounts. The derivation of the estimated errors in transmission is described in Appendix D. A mean absorption spectrum for the nine days of observations was also produced by averaging all the individual spectra together. The averaged absorption predicted by the monomer theory was then subtracted from the observation leaving the average excess absorption contour shown in Figure 6.4. Two other experimental measurements of excess absorption have been included on Figure 6.4. These were scaled to slant path atmospheric conditions using Eqn.(2.9) which assumes the excess absorption was caused by the water vapour dimer. The excess absorption has not been conclusively proved to scale according to 2.9, so that the following comparisons are only strictly valid if dimers cause the discrepancy between monomer theory and observation.

The slant path excess absorption was on average greater in magnitude than that observed under controlled conditions in the laboratory by
Fig. 6.3 Daily averages of transmission for three days with different water amounts. The number of individual spectra in each daily mean varied between 9 and 15. The dashed spectrum is the monomer prediction and the dot-dashed spectrum is the monomer with the equilibrium dimer prediction included. The error bars include both instrumental noise and uncertainties in the calibration.
Fig. 6.4 A comparison of the mean absorption observed in excess of the monomer prediction in the laboratory (dashed line), through a slant path (solid line) and an open air horizontal path (squares). The error bars for the slant path measurement indicate the experimental uncertainty, for the horizontal path they indicate the variability observed about the mean value.
Bohlander (1979). Measurements of excess absorption taken in an open air white cell (Emery et al. 1979) are also plotted showing that it was on average greater in a horizontal path than in a slant path.

Figure 6.5 compares the observed mean transmission spectrum with that predicted by Gaut and Reifenstein (1971) derived using their monomer theory and adding the correction factor defined by Eqn. (2.11). The mean radiosonde profile was used to calculate this transmission spectrum. There was good agreement between the observed slant path transmission and that predicted by this technique.

6.4 Anomalous Absorption Features Between 6.5 and 10 cm⁻¹

In the window region between 6.1 and 10.8 cm⁻¹ anomalous absorption features were often seen that were consistent over short time periods. Figure 6.3(a) shows a definite increase in absorption at 7.7 cm⁻¹ in the daily average spectrum, which was also seen on the following day with about the same amount of water vapour in the path. Figures 5.3(b) and (c) show similar discrete absorptions for higher water amounts but their frequencies have shifted. These features were not always present, however, and they were not simply correlated with water amount or presence of cloud. The fact that the features do change frequency and sometimes disappear altogether makes it less probable that they were instrumental in origin. An example of their consistency over short time periods is shown in Figure 6.6, the three individual spectra plotted were all recorded within an hour of each other. They all exhibit a dip at 7.5 cm⁻¹, whereas the spectra taken an hour later were much smoother, suggesting that a change had occurred during that time period.

These anomalous features were also evident in the spectrum shown in Figure 6.7(a) taken from Imperial College and in 6.7(b) taken from Queen...
Fig. 6.5 A comparison between the mean observed transmission spectrum (solid line) and that predicted from the Gaut and Reifenstein formulation (dashed line).
Fig. 6.6  Three individual spectra recorded consecutively over a period of an hour. The zero level for each spectrum is shifted to separate them for clarity, the first spectrum is the lowest.
Fig. 6.7  (A) A daily mean transmission spectrum recorded from Imperial College with a Golay detector. (B) A mean transmission recorded by Harries and Ade (1972) at Queen Mary College, London. (C) An overall mean transmission spectrum of the observations taken from Mauna Kea, Hawaii (Moffat et al. 1977).
Mary College, London by Harries and Ade (1972). They also appeared in spectra taken from high altitudes. An overall mean transmission spectrum taken from Mauna Kea, Hawaii (Moffat et al. 1977) is shown in Figure 6.7(c) which has a pronounced dip at 7.7 cm\(^{-1}\).

Features were seen in the 4-6 cm\(^{-1}\) window, but the random errors were much larger in this region so that no real significance could be attached to them.

### 6.5 The Absorption at 7.7 cm\(^{-1}\)

A few measurements of atmospheric absorption at 7.7 cm\(^{-1}\) have been made from sea level using radiometer techniques, as described in Section 1.3. The measured transmission at 7.7 cm\(^{-1}\) was converted to absorption coefficient for every individual spectrum and then plotted against precipitable water, as shown in Figure 6.8. The total absorption was correlated with water amount and was significantly greater than the monomer theory predicts. There also appeared to be a certain amount of variability in the absorption values, which was caused by the anomalous absorption features mentioned in the previous section. The best fit straight line to the observations which was forced to go through the origin was

\[
A(dB) = 0.33 \omega \quad \text{... (6.6)}
\]

where \(\omega\) is in millimetres precipitable in the range from 5-25 mmppt.

The standard deviation of the slope was ±0.08. The absorption values below 10 mmppt were all above the line because of a persistent anomalous feature at 7.7 cm\(^{-1}\) seen on both days with low water amounts.

Wrixon and McMillan (1978) found absorption values on average somewhat less than our measurements, although on one day our measurements were close to their fitted line. The measurements taken by Plambeck (1978) show good agreement (assuming a scale height of 2 km for water vapour) and
Fig. 6.8 The absorption at 7.7 cm⁻¹ (crosses) as a function of precipitable water in the path. The solid line is the monomer prediction using the Gross line shape, the dashed line was the best fit line through the observations and the origin. The diamond marks the measurement made by Ulaby and Straiton (1969). The error bars indicate the experimental uncertainty of each point.
his observations also show considerable variability, although this may be accounted for by clouds, rain or snow. The measurements of Johnson et al. (1970) were in agreement, the best fit line through their results was $A(dB) = 0.28\omega$ at 7.7 cm$^{-1}$.

6.6 Attenuation by Cloud

The solar and sky emission spectra were only recorded when the sky was clear or when the cloud present had a negligible effect on the broad band signal. However, the effects of various types of cloud on the signal were investigated. Figure 6.9(a) compares the attenuation at near infrared wavelengths (1.2 $\mu$m) with far infrared wavelengths (3–11 cm$^{-1}$) through a thin layer of cirrus cloud. The far infrared signal was unaffected by cirrus, but there were definite fluctuations in the near infrared signal. Figure 6.9(b) shows the effect that thick cumulus clouds, cumulus congestus, have at both wavelengths. The near infrared signal was totally absorbed during the passage of a cumulus cloud across the sun, whereas the far infrared signal was only attenuated by 3 to 6 dB. The smaller 'fair weather' cumulus had very little effect at millimetre wavelengths.

The far infrared signal occasionally underwent fluctuations when the near infrared signal remained constant, as indicated by the arrow in Figure 6.9(b). This may have been due to a sudden decrease in the amount of water vapour in the path (the near infrared signal was not in a water vapour absorption band so it was insensitive to the amount of water in the path). When cumulus clouds were forming it would not be surprising if the water vapour in the path was varying appreciably. Greve (1978) saw similar variations in water vapour amount over time periods of minutes measured by a near infrared hygrometer. Apart from these slow fluctuations over time periods of minutes, the noise level of the solar signal was no more than that measured when looking at the black body source in the laboratory.
Fig. 6.9 The effect of cirrus clouds (A) and cumulus congestus clouds (B) on near and far infrared signals. The vertical arrow in (B) indicates a change in the far infrared signal not caused by cloud.
At millimetre wavelengths scattering by cloud droplets which are usually less than 50 μm in diameter can be calculated using Rayleigh scattering theory. The predicted scattering coefficient was calculated to be 11 dB/km/g/m³ at 7 cm⁻¹. Mason (1971) gave values for the mean water content for various types of cloud and quoted a value around 0.4 g/m³ for cumulus clouds which would give a predicted attenuation of 4.4 dB.
CHAPTER 7 - APPLICATIONS AND CONCLUSIONS

7.1 Applications

7.1.1 Prediction of Atmospheric Absorption above any Height

The atmospheric absorption above a site at any elevation can be calculated using the appropriate version of the U.S. Standard Atmosphere (1966). Figure 7.1(a) shows the average precipitable water above a site as a function of height calculated from the temperate winter and summer model atmospheres. The vertical water amount derived near sea level was higher (30 mmppt) in the summer than the values commonly observed (20 mmppt). This was because the measurements were only taken on dry, cloud-free days, whereas the model atmosphere included cloudy days in the average.

The monomer absorption depends upon the broadening conditions above a site as well as the water amount, so that Eqn. (6.6) relating precipitable water to the absorption derived from observations will only be strictly correct for observations from sea level. Figure 7.1(b) plots the predicted absorption averaged over the 7-10 cm⁻¹ window as a function of water vapour amount with the broadening conditions varying with the water amount. This was achieved by using Figure 7.1(a) to give the height corresponding to a particular water amount and then calculate the mean broadening conditions above that height using Eqn. (2.10). In fact the broadening parameters have very little effect, so that the monomer absorption is very nearly linearly related to water amount. However, the broadening conditions also change with season. The predicted monomer absorption shown in Figure 7.1(b) was derived for the summer months. During the winter months the total absorption was about half a decibel above the line shown in Figure 7.1(b) at low altitudes. The dimer component was calculated for the model atmosphere conditions as described in Section 2.5.

In practice the total absorption observed was usually greater than
Fig. 7.1 Model atmosphere predictions for (A) precipitable water above a particular height (temperate latitudes) and (B) absorption predicted from the AFCRL tabulation during the summer, with the broadening conditions decreasing for low water amounts to simulate conditions above a mountain site.
The uncertainty involved in predicting the atmospheric absorption at 7.7 cm\(^{-1}\) could be large, because of transient absorption features which appeared from time to time. Figure 6.8 illustrates that if Eqn. (6.6) was used to predict the absorption at low altitudes the value deduced can differ by as much as 2 dB from the observation. This variability may explain why some high altitude sites investigated had low anomalous absorption values, whilst others had much higher values.

7.1.2 Remote Sensing at Millimetre Wavelengths

At the present time most of the remote sensing of the atmosphere from space is carried out in the visible and mid-infrared wavelengths up to 20 \(\mu\)m. However, at these wavelengths cloud significantly effects the signal whereas millimetre wavelengths have the advantage that they are only strongly attenuated by dense water clouds, as shown in Section 6.6.

Temperature profiles of the atmosphere could be obtained by scanning over the strong oxygen band at 2 cm\(^{-1}\) as demonstrated by Waters et al. (1975). Similarly as techniques improve it may prove possible to obtain water vapour profiles using the 6.1 cm\(^{-1}\) water line if varying anomalous absorption features do not exist in this region.

Images of the earth at millimetre wavelengths would be of interest to the meteorologist for probing the internal structure of intense depressions and hurricanes. The thinner clouds around the edge of the depression and the cirrus clouds above would be essentially transparent at millimetre wavelengths, allowing the active areas of depressions and hurricanes to be observed more easily. These kind of images may also help to probe other planetary atmospheres such as Venus and Jupiter, and aid our understanding of the cloud structures seen at shorter wavelengths.
7.1.3 Communications at Millimetre Wavelengths

As the lower frequency regions become more congested the possibility of communicating at higher frequencies becomes increasingly more attractive. In the 7-10 cm\(^{-1}\) window the atmospheric absorption is at least 2 dB/km (assuming no anomalous absorption), so that communication would be feasible only over short distances (~few kms). This could have advantages in a city where many short communication links could operate at the same frequency with no interference as long as they were a few kilometres apart.

An obvious military application would be that information could be transmitted fairly close to the enemy without fear of being intercepted because of atmospheric absorption. These systems would be strongly attenuated by rain and any anomalous absorption present would cause fading. More research would be needed to ascertain how important these restrictions are.

7.2 Possible Mechanisms for Anomalous Absorption

The anomalous absorption observed between 6 and 10 cm\(^{-1}\) consists of variable absorption features which were not simply correlated with any obvious atmospheric variable. They were not in a steady state since features were observed to appear and disappear over a few hours.

When these features were originally seen by Harries and Ade (1972) it was suggested by Kislyakov (1972) that they were caused by absorption lines of minor atmospheric constituents such as O\(_3\), CO, NO and N\(_2\)O. However, both Harries (1972) and Gimmestad et al. (1972) carried out calculations to show that the features from these minor constituents would be negligible in comparison with those observed. Ozone was the only constituent with a significant absorption and this was included in the theoretical spectra described in Chapter 2.
Harries and Ade (1972) assigned one of the features they observed to a Q-branch line of the water vapour dimer rotational spectrum at 7.7 cm\(^{-1}\) which was calculated by Viktorova and Zhevakin (1971). However, more recently Viktorova and Zhevakin (1973) have stated that according to their calculations the Q-branch line would only be detectable if the concentration of dimers was one or two orders of magnitude greater than that expected in the atmosphere under equilibrium conditions.

The higher anomalous absorption observed in horizontal paths suggests that the phenomenon is enhanced near the ground.

Possible mechanisms proposed for this anomalous absorption are highly speculative. Apart from dimers, other polymers of water vapour might be expected to contribute some absorption at these wavelengths. The presence of an exposed liquid water surface may increase the absorption across horizontal paths, if water vapour polymers "evaporating" from the surface traverse the line of observation.

Theoretical studies have been extensively carried out on the scattering of electromagnetic radiation by water droplets and the absorption of radiation by individual water molecules, and dimers. However, aggregates of a few hundred water molecules condensed on salt nuclei may also exist in the atmosphere, which are too small to constitute a cloud droplet and yet too large to be a simple polymer. These "embryo drops" would not be in equilibrium with the surrounding vapour but they may exist for long enough to have some effect. If they were polarised in some way they could interact with the radiation field around them, causing anomalous features at millimetre wavelengths.

### 7.3 Conclusions

The absolute transmission of the atmosphere through a slant path from sea level has been measured in the 3 to 15 wavenumber range. The strong
water vapour and oxygen absorption lines observed, showed good agreement with the predicted frequencies and strengths given in the A.F.C.R.L. listing (McClatchey et al. 1973).

The window regions between the lines absorbed twice as much radiation on average than was predicted by the Gross line shape. If an estimate of the water vapour dimer absorption derived from laboratory measurements is also included the discrepancy between theory and prediction is less but a variable component of anomalous absorption still remains. The mechanisms causing this absorption are not understood at the present time.

The absorption at 7.7 cm\(^{-1}\) (in the middle of a window region) was found to be correlated with water amount and air mass, although a good deal of variability about the mean was observed. The mean absorption from sea level at 7.7 cm\(^{-1}\) was given by the relation \(A(\Delta B) = 0.33 \omega\) where \(\omega\) is in millimetres precipitable. The variability was caused by transient anomalous absorption features which did not appear to be strongly correlated with any obvious atmospheric variable.

The excess absorption observed in a horizontal path was greater than seen in slant paths suggesting that anomalous absorption is enhanced close to the surface.

The water amount in a slant path was obtained from water vapour absorption bands in the near infrared region. The bands were calibrated in two ways, the first was to calculate spectra from the A.F.C.R.L. listing and then relate the integrated absorption of the bands to precipitable water. The other method utilised water amounts obtained from a radiosonde to calibrate the observed integrated absorptions. A fifty per cent discrepancy was found to exist between the radiosonde and A.F.C.R.L. water amounts for the 1.38 \(\mu\)m band but better agreement was obtained for the 0.94 \(\mu\)m and 1.14 \(\mu\)m bands. Very recently an updated version of the A.F.C.R.L. listing has become available (Rothman, 1978) which may reduce this discrepancy.
7.4 Suggestions for Further Work

To investigate the discrepancies between the observed integrated absorptions of the near infrared water vapour bands and those predicted by the A.F.C.R.L. listing, a horizontal path experiment similar to the one described by Emery et al. (1979) but using all three bands could be carried out. Measurements both under controlled conditions and through the real atmosphere would be useful to ascertain whether the discrepancies arise from an atmospheric phenomenon or deficiencies in the A.F.C.R.L. listing.

In the far infrared region it appears that anomalous absorption is enhanced close to the ground, which suggests that open air horizontal path experiments would be the best method to investigate this phenomenon. Meteorological variables such as the presence of a liquid water surface, wind speed and visibility, may all prove to be important factors. It would be particularly interesting to carry out an open air transmission experiment at lower temperatures where the dimer absorption is predicted to increase.

Simultaneous measurements of attenuation at millimetre wavelengths and at 10 μm would be useful as anomalous absorption is present in both wavelength regions. The main objective would be to determine if the intensity of anomalous absorption was correlated in both windows.

Transmission measurements of the atmosphere over equatorial regions can now be systematically made using geosynchronous meteorological satellites, which measure the intensity of terrestrial radiation emitted by the ground and atmosphere at 11 μm. Knowing the temperature and water vapour density profiles for the region which is emitting the radiation, the atmospheric transmission from earth to space can be inferred. Using the A.F.C.R.L. listing it should also be possible to calculate a monomer transmission value to ascertain the amount of excess absorption present. The advantage of using this technique is that the atmosphere in equatorial
regions has appreciably higher water vapour densities at the surface than at temperate latitudes, which may increase the anomalous absorption.
A.1 Introduction

An implicit assumption in the black body calibration is that the telescope mirror is completely filled with radiation from all points of the solar disc as is the case with the black body disc. For large hour angles during the summer months the angle between the sun and the meridian is large, causing one axis of the heliostat mirror to be considerably reduced when viewed from the telescope. When this minor axis $BB'$ in Figure 3.3 is reduced to such an extent that $AA'$ is smaller than the telescope mirror then a correction has to be applied because radiation from all points on the sun will not be able to reach the edges of it. The following section describes the method used to correct for this.

A.2 The Correction

If you place your eye in the plane of the telescope mirror $AA'$ and then look at the heliostat mirror, the whole solar disc should be visible but as you move your eye towards the edge of the telescope mirror there will become some point where the observed solar disc touches the edge of the heliostat mirror. The problem is to estimate how the intensity falls off as the solar disc disappears from view at the edge of the heliostat mirror.

The solar disc is circular whereas the heliostat mirror boundary is an ellipse. The solar intensity at a certain distance $d$ from the centre will be proportional to the area shaded in Figure A.1(a). To calculate this area, the points of intersection of the circle and ellipse were determined and then the areas under the circle and ellipse can be derived numerically:
Fig. A.1 (A) The solar image as seen from the telescope mirror when the heliostat mirror begins to obscure it. (B) The intensity deduced from the shaded area in (A) as a function of distance from the optical axis. The heliostat mirror was 40.5 cm diameter, the telescope mirror 30.5 cm and their distance apart 10 m.
Area = \[ 2 \int_{d-r}^{x_1} \left( \frac{x^2 - (x-d)^2}{2b} \right)^{1/2} dx + 2b \int_{x_1}^{a} \left( 1 - \frac{x^2}{a^2} \right)^{1/2} dx \] ... A.1

where \( x_1 \) = point of intersection
\( a, b \) = semi-minor and major axes of the ellipse
\( r \) = apparent radius of solar disc

This area was determined for a range of values of \( d \) to give the intensity distribution along one axis across the telescope mirror, as shown in Figure A.1(b).

To calculate the total power reflected from the mirror, its surface was divided up into "concentric" ellipses with the innermost marking the boundary where the sun begins to be obscured. The values of the intensity in the regions between each ellipse decreased according to the curve in Figure A.1(b).

The intensity multiplied by the corresponding area overlapping the telescope mirror was then integrated over the mirror's surface to give the total power reflected. If the complete solar disc was visible from every point on the surface, the total power would be proportional to the area of the mirror's surface. The calculated power was divided by this to give a correction factor (less than unity) which was applied to the observed solar intensity.

For the observations described in this thesis the correction factor never differed from unity by more than 6%.
APPENDIX B - SCREW ERRORS

The Michelson Interferometer consists of two plane mirrors at right angles to each other. One is fixed whilst the other moves in the direction perpendicular to its surface, to alter the phase of the radiation at the beamsplitter where it is recombined with the radiation from the fixed mirror. The movement of this mirror was controlled by a stepper motor which rotated a screw thread that drove the mirror in the required direction. The mirror was moved 26.4 μm between each sample which gave a cut off frequency of 94.6 cm⁻¹. If this distance varied due to periodic errors in the screw thread, then the resultant spectrum would be distorted.

The screw thread was checked for errors by using an accurate micrometer probe that could measure the distance the mirror moved to an accuracy of one micron. It was found that the thread was slightly distorted which periodically caused the mirror to be moved by 8% more or less than the mean distance. This cyclic pitch error can introduce 'ghosts' into the spectrum if sharp lines are present. If the mirror movement for one revolution of the screw thread is X cm, then if cyclic errors exist, 'ghosts' would be expected at ±\( \frac{1}{4X} \) cm⁻¹ from a sharp feature in the spectrum (Connes, 1963). The intensity of the spurious features will depend upon the magnitude of the screw error.

To investigate whether the measured error of 8% introduced any artefacts into the transmission spectrum, the amplitude and phase of the pitch error with respect to zero path difference was measured. An original interferogram was then resampled using an interpolation programme so that each point of the interferogram represented the value that should have been recorded if there had been no screw error. A solar and background interferogram were corrected in this manner to give a corrected transmission spectrum. This was compared with the uncorrected spectrum and the difference at millimetre wavelengths was found to be negligible.
At shorter wavelengths the errors in the screw pitch will have more effect. The above correction procedure has been successfully applied to interferograms taken at ultra-violet wavelengths (Bohlander, 1979) to remove much of the distortion in the spectrum caused by pitch errors.
APPENDIX C - DIFFRACTION EFFECTS

C.1 Theory

At visible wavelengths the circular stop in front of the interferometer was filled with either solar or black body radiation of uniform brightness temperature. At millimetre wavelengths diffraction becomes significant because as the wavelength approaches the dimensions of the mirror, the focussed image becomes broadened due to Fraunhofer diffraction. A point source infinitely far away from a circular telescope mirror will give an intensity distribution in the focal plane (Born and Wolf, 1964) of:

\[ I(r) = 4 \left( \frac{J_1 \left( \frac{\pi Dr}{\lambda f} \right)}{\frac{\pi Dr}{\lambda f}} \right)^2 \]  ... C.1

where  
- \( J_1 \) = the Bessel function of the first order  
- \( D \) = diameter of telescope mirror  
- \( f \) = focal length of the mirror  
- \( r \) = distance from the centre of the focal plane  
- \( \lambda \) = wavelength of radiation

In order to calculate what effect diffraction has on the solar image it is necessary to convolve the undiffracted image in the focal plane with Eqn. C.1.

A simple one dimensional approach to this convolution is to assume that the undiffracted image is a top hat function, and the diffraction function to be one dimensional, as given by Eqn. C.1. These two functions can then be convolved together to give the diffracted image. This one-
dimensional calculation is an approximation since the solar image is a disc not an infinitely long strip, as assumed in the above analysis.

Because of the symmetry of the circle it is possible to perform a two-dimensional convolution along one diameter of the undiffracted solar or black body disc. If the Airy disc (the diffraction function in Eqn. C.1 rotated $360^\circ$ about $r = 0$) is placed close to the undiffracted image, the total intensity $I(Z)$ at a distance $Z$ from the centre of the image can be calculated by dividing the Airy disc up into annuli of successively larger radii. The intensity of the source surround was assumed to be zero for the solar image (sky emission was removed in the transmission spectra). Whenever an annulus overlays the solar image, as shown by the shaded region in the diagram, the area of this region is calculated and multiplied by the solar intensity and the appropriate value of the Airy disc at that radius. To calculate $I(Z)$ there are three regions to consider:
(i) where the annulus is not big enough to touch the image \\
\( r < Z - R \)

(ii) where the annulus and image overlap \( Z - R < r < Z + R \)

(iii) where the annulus is too large to touch the image \( r > Z + R \)

As the surround intensity is zero, only the radiation from the overlap in (ii) contributes to \( I(Z) \) so the integral becomes:

\[
I(Z) = \left[ \int_{Z-R}^{Z+R} \left( \frac{J_z(\rho)}{\rho} \right)^2 I_0 2r d\theta dr \right] 
\]

where \( \rho = \frac{\pi DR}{\lambda f} \) as defined in C.1

\[
\theta = \frac{\pi}{180} \cos^{-1} \left( \frac{Z^2 + r^2 - R^2}{2Zr} \right) \text{ radians}
\]

The above integral was evaluated numerically for values of \( Z \) from the centre of the disc out to a distance where the value of \( I(Z) \) had become essentially zero. The resultant function \( I(Z) \) gives the intensity of the diffracted image along one axis in the focal plane for a particular wavelength. Figure C.1 shows a plot of the undiffracted and diffracted images in the focal plane for a wavelength of 1.43 mm (7 cm\(^{-1}\)).

To allow for the warm surround of the black body an annular region around the circular image was included which had an intensity of 62% of the central disc and outer diameter 1.62 times larger than the disc. These figures were derived from thermocouple measurements of the temperature close to the black body aperture. To calculate \( I(Z) \) the same type of analysis as described above can be used because the circular symmetry is still present, although there are more regions to integrate over in this case.

The edges of the entrance stop are indicated in Figure C.1. For the calibration to be correct the solar and black body diffracted images should be identical inside these limits.
Fig. C.1 The predicted intensity of solar radiation across the aperture stop at visible wavelengths (solid line) and at 7 cm\(^{-1}\), dashed line. Also included is the black body intensity at 7 cm\(^{-1}\), the effect of the warm surround is clearly shown.
Fig.C.2  The ratio of solar over black body power passing through the stop according to Fraunhofer diffraction theory.
The total power entering the interferometer at a particular wavelength for a circular stop is given by

\[ W(\lambda) = 2\pi \int_0^s I(z) z \, dz \]

where \( s \) = the radius of the stop
\( z \) = the distance from the centre of the stop

The ratio of the power from the sun over the power from the black body for various wavelengths gives the correction curve shown in Figure C.2. This illustrates that more black body radiation passes through the stop at longer wavelengths than solar radiation because of the warm black body surround. This correction could be applied to the transmission spectrum if all of the radiation passing through the stop was detected. To investigate if this assumption was valid a few experimental checks were carried out (Section C.2).

Fresnel diffraction was also considered as both the heliostat mirror, for the solar radiation and the moving mirror for the black body radiation would spread out the beam. The difference between the two paths of radiation was that the heliostat mirror was over twice as far away from the telescope compared with the movable mirror (see Figure 3.2). By assuming the mirrors to be square, the formulae given by Goodman (1968) were used, to obtain the intensity distribution in the plane of the telescope mirror for both sources. The differences found at 7 cm\(^{-1}\) were not appreciable although at longer wavelengths this would become more important.

**C.2 Experimental**

The Fraunhoffer diffraction problem was investigated experimentally by looking at three sources with and without the stop in front of the interferometer. To determine what effect the aperture stop had on the
spectrum, a source with a uniform intensity distribution (liquid nitrogen surface) was used. The result shown in Figure C.3 which is a ratio of the spectrum taken through the stop over the spectrum with no stop, shows that the stop acts like a high pass filter strongly attenuating all longer wavelengths.

The corresponding ratios for the black body and solar images are similar in shape as shown in Figure C.3 when the ratios are normalised to unity for short wavelengths. The solar image (with sky emission removed) would be expected to be more strongly attenuated at longer wavelengths, as indicated by the theory above. In fact the observations show very little difference between the black body and solar ratio. This suggests that although the black body surround would have leaked through the stop, the radiation near the edge of the stop was not all detected causing the theoretical correction curve to be not applicable.

The transmission spectra were not corrected for diffraction effects because experimentally the effect of the stop appeared to be the same for either source. Further evidence that diffraction effects were negligible was that the calibrated transmission spectra were in good agreement at all wavelengths with the zenith angle spectra which were not subject to diffraction effects (see Figure 6.2).

C.3 Conclusions

The uncertainties which arise at millimetre wavelengths due to Fraunhoffer diffraction effects can be reduced by using an aperture stop small enough so that radiation surrounding the image cannot enter the interferometer.

Bradsell and Harries (1978) have suggested that one way to decrease the side lobes of the intensity distribution of Eqn.C.1 is to employ an apodizing technique. This could be achieved by slowly reducing the reflectivity of the telescope primary at the edge of the mirror.
Fig. C.3 The effect of placing the aperture stop in front of three different intensity distributions. (A) shows a source of uniform intensity (liquid nitrogen surface), (B) the sun and (C) the black body centred on the stop. All ratios were normalised to unity at short wavelengths.
Uncertainties due to Fresnel diffraction could be reduced by arranging that the black body radiation is incident upon the heliostat mirror which then reflects the radiation on to the telescope.
APPENDIX D - ERROR ANALYSIS

D.1 Random Transmission Errors

The signal to noise ratio of the system was limited by detector noise for both the solar and black body spectra. Any interferograms which had numerous spikes or significant gain changes caused by clouds were rejected.

After checking that no significant gain or spectral changes had occurred during the day, the uncertainty in the constant background spectrum for that day was obtained by taking the root mean square (R.M.S.) deviation of each individual spectra from the average spectrum:

\[ \Delta B_{RMS} (v) = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left( B_i (v) - B (v) \right)^2} \]  ... D.1

where \( B_i (v) \) = the \( i \)'th individual spectrum
\( B (v) \) = the average spectrum
\( N \) = the number of individual spectra.

This gives the uncertainty for one individual background spectrum, the uncertainty in the average background is then given by:

\[ \Delta B (v) = \frac{\Delta B_{RMS} (v)}{\sqrt{N-1}} \]  ... D.2

where \( N \) is the number of spectra in the average

The solar spectra varied during the day so the above analysis cannot be applied. To calculate the uncertainty in the solar spectrum \( \Delta S (v) \) the broad band solar intensity was recorded for a constant path difference well away from zero. For an amplitude modulated interferogram this signal will be the average intensity recorded during an interferogram. This method will include any sky noise present in the estimate of \( \Delta S (v) \).
The signal was recorded for the same length of time as a normal interferogram and was then Fourier transformed to produce a noise spectrum which was squared and square rooted to give $\Delta S_{RMS}(v)$. In calculating the transmission spectrum the solar spectrum has the sky emission spectrum subtracted so that the uncertainty becomes $\sqrt{2} \Delta S_{RMS}(v)$.

Knowing $\Delta S(v)$ and $\Delta B(v)$ the transmission uncertainty $\Delta t(v)$ for an individual spectrum can be found by differentiating Eqn.(4.4) and taking the RMS value

$$\Delta t_{RMS}(v) = \frac{K(v)}{B(v)} \left[ 2\Delta S^2(v) + \frac{t^2(v)}{K^2(v)} \Delta B^2(v) \right]^{\frac{1}{2}}$$

... D.3

where $K(v)$ = the ratio of temperature differences in Eqn.(4.4)

$t(v)$ = a smoothed average transmission for the particular day

$N$ = number of spectrum in the average background

This uncertainty can be reduced by averaging transmission spectra taken on the same day.

To check that the method of obtaining $\Delta S(v)$ was giving a realistic estimate of its value, the same method was used to estimate $\Delta B(v)$ and this was then compared with the value derived using Eqn.D.1. Figure D.1 shows the two values for $\Delta B(v)$ found by the different methods, in the 5-10 cm$^{-1}$ region they are about the same. The value of $\Delta F(v)$ for the same day is also plotted, showing that the atmosphere is not contributing a significant amount of noise to the signal at these wavelengths.

An error analysis for individual background spectra divided by an average background spectrum can also be derived to give the uncertainties shown in Figure 4.2.

The RMS error from unity, $\Delta R$, is given by
Fig. D.1 Three noise spectra. The solid line is the solar noise spectrum produced by Fourier transforming the solar signal at a constant path difference. The dashed line is the black body noise determined in the same way and the dot-dashed line is the black body noise determined by taking the R.M.S. difference of individual background spectra from the daily mean.
\[ \Delta R(v) = \frac{\Delta B(v)}{B(v)} \left[ 1 + \frac{1}{N-1} \right]^{\frac{1}{2}} \] ... D.4

where \( N \) = the number of backgrounds in the average.

A similar analysis can be used to calculate the uncertainties in the zenith angle spectra.

D.2 Consistent Errors in Transmission

The errors described above represent the differences that arise between two identical transmission spectra because of the limiting detector noise. A consistent error inherent in every transmission spectrum may also be present. To determine the ratio of the temperature differences in Eqn. (4.4) the brightness temperatures of the sun \( T_s(\text{°K}) \) and black body \( T_B(\text{°K}) \) have to be known. The uncertainty in solar temperature is estimated to be ± 400 (°K), (Bastin et al. 1964) and in the black body temperature to be ± 10 (°K), which gives an uncertainty of ± 6.5% of the transmission value. Uncertainties in the calibration due to diffraction discussed in Appendix C increase with wavelength. The total uncertainty at 5 cm\(^{-1}\) was estimated to be ± 20% whereas at 8 cm\(^{-1}\) it was only ± 8%.

D.3 Errors in Precipitable Water Measurements

The uncertainty in measured water amount was obtained by plotting water amounts obtained from one band against water amounts from an adjacent band measured at the same time, shown in Figure D.2. The spread of values about the mean line where both bands are in agreement, gives an estimate of the random error associated with a single measurement. A random error of 10% in the water amount was estimated.
Assuming the radiosonde water values were on average correct (individual radiosonde values can be in error by as much as 16% (Saiedy, 1960)) and the precipitable water above Slough was on average the same as at Crawley, there should be no consistent error in the water amount derived.

The predicted absorption in the window regions will be proportional to water amount (neglecting \( O_2 \) and \( O_3 \) absorption) which gives a 10% uncertainty in the absorption predicted by the monomer theory. This uncertainty has to be included with the measured uncertainties, when a value for the uncertainty in the excess absorption is estimated.
Fig.D.2 Water amounts obtained almost simultaneously from adjacent near infrared bands. The spread of values about the straight line gives an idea of the random error of an individual measurement.
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