RECENT PROGRESS IN INFRARED AND MICROWAVE ATMOSPHERIC SOUNDING

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RECENT PROGRESS IN INFRARED AND MICROWAVE ATMOSPHERIC SOUNDING

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1 INTRODUCTION

As implied in the title, the purpose of this paper is to provide a report on recent progress in atmospheric spectroscopy in the infrared and microwave regions, and also by implication the intermediate spectral range namely the far infrared. We arbitrarily define these regions in terms of wavelength as 1 - 30 μm, 30 - 500 μm and 500 μm - 5 mm.

We shall not attempt a review of the historical background to the subject; nor shall we give details of the basic methods used in atmospheric spectroscopy and their advantages and disadvantages, since this aspect was dealt with in a recent paper.1

The application if IR techniques to studies of the Earth's atmosphere is not recent, and considerable work has been done over a number of years to develop such methods.2 Applications of submillimetre3 and microwave4 methods, however, is more recent, having been strongly developed in only the past decade. As we shall see below each of these quite distinct spectral regions has the potential to make significant contributions to atmospheric science.

The paper will be organised as follows: Section 2 will list the types of instrument and technique used in current research. Sections 3, 4 and 5 will then review some of the most recent progress in the infrared, submillimetre and microwave regions respectively. Section 6 will provide some comments on likely future developments.

2 TECHNIQUES

A wide variety of instruments and techniques is available to the modern atmospheric spectroscopist. A short review of available techniques has been presented elsewhere, and for present purposes we shall merely summarise, by means of Table 1, the techniques currently being used by several groups active in this field. An explanation of the entries may be found in the key attached to the table. We should only note here that a wide variety of methods, including gas correlation spectroscopy, resonance fluorescence spectroscopy, absorption and emission spectroscopy, implemented through several different types of instruments are now in use. Bearing in mind that Table 1 is almost certainly incomplete, there is clearly a great deal of activity in atmospheric spectroscopy at a number of institutions. Some of the more outstanding results obtained by this community in recent times will be reviewed below.

3 RECENT RESULTS IN INFRARED SPECTROSCOPY

Very considerable advances in our understanding of the details of the IR spectrum of the atmosphere have been made through the work of D G Murcray, A Goldman, J Williams and their colleagues at Denver University. Perhaps the most famous and appealing result was that obtained about a decade ago, when a balloon-borne grating spectrometer was used in solar occultation configuration to make measurements of the absorption spectrum in the 11 μm region over very long optical paths. As is illustrated by Figure 1, as measurements were made at increasing zenith angles from about 89° to 94°, a quite strong absorption band (the v5 and 2v5 vibrations) due to HNO3 was revealed. The nitric acid molecule had previously been identified at different wavelengths (v4 band at about 1300 cm⁻¹) by this group, but the result at 11 μm provided a very clear and convincing confirmation of the existence of HNO3, in a layer centred at about 25 km altitude, in the stratosphere. The v5 and 2v5 bands have since been used extensively by a number of groups for remote measurements of this atmospheric constituent.

1 Measurements of solar transmission in the 800-950 cm⁻¹ region from a balloon floating at approximately 30 km. Each trace is a separate spectral scan, obtained at the solar zenith angle shown against it, and the results are displaced by 20% for clarity. The growth of absorption features due to HNO3 are clearly observed. (From ref.5)

* Written in part while the author was on leave of absence at the National Center for Atmospheric Research, Boulder, Colorado, USA.
Another example of the very clear and methodical work which has been done by the Denver group in order to clarify the details of the IR spectrum is shown in Figure 2. This example also illustrates the importance of a thorough species-by-species consideration of a complex atmospheric spectrum when trying to assign features observed in that spectrum; the most convincing way of achieving a firm set of assignments is to build up an exact simulation of the observed spectrum, using either measured laboratory spectra for the different gases involved, or computer simulation of their spectra using theoretical models of the molecular absorption processes. Figure 2 shows a solar transmission spectrum in the 1602-1618 cm⁻¹ region (Frame 1) obtained from an aircraft flying at ~15 km. Frames 3, 4 and 5 show calculated spectra for three molecules H₂O, NO₂ and CH₄ respectively and the second frame shows the product of 3, 4 and 5. The close similarity between the top two frames clearly shows that NO₂ absorption features have been detected (eg at 1604.5 cm⁻¹, 1606 cm⁻¹ and 1611.5 cm⁻¹), and also that all the major features of this spectral region are well understood. Some details remain to be clarified though (of the small features at 1613.0 - 1613.5 cm⁻¹), and one suspects that remaining uncertainties may well reside in imperfect spectral line data.

Extensive measurements of the IR absorption spectrum have also been made for several years by the group at the Jet Propulsion Laboratory under C B Farmer. The technique used by this group is high speed (of scanning) Fourier transform spectroscopy using a Michelson interferometer. An example(7) of the work of this group is given in Figure 3, which shows portions of atmospheric absorption spectra in two spectral regions, around 2926 and 4038 cm⁻¹, observed using solar occultation methods from a balloon at ~35 km altitude. At these wavelengths thermal emission intensities are very weak at atmospheric temperatures and solar absorption offers the only practicable method of making measurements of minor constituents. At the wavelengths shown, the spectrum in Figure 3 shows absorption features due to telluric HCl and HF (from which profiles of the HCl and HF distributions were calculated) as well as solar CO (marked 8). Others (including Zander(8) et al and Buiga(9)) have also made measurements at these short IR wavelengths, using high resolution grating spectrometers or Michelson interferometers mounted on balloons.

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2 Measurements of solar transmission in the 1610 cm⁻¹ region from an aircraft at about 15km altitude (Frame 1), compared with a calculated spectrum (Frame 2) which is produced from the individual spectra shown in Frames 3 - 5. (From Ref. 21)

3 Atmospheric transmission of solar radiation measured along a limb path at an altitude of about 35 km using a Michelson interferometer in the 2900 and 4000 cm⁻¹ regions. Absorption features due to stratospheric HF and HCl, and solar CO (9) are marked. (From Ref. 7)
In Europe, high resolution IR spectroscopy has been studied in detail by the groups under Girard in France and Ackerman in Belgium, often working together.[10]. Measurements made by this group have utilised the ingenious Grille spectrometer[11] which operates essentially as a grating spectrometer but utilises compound entrance and exit slits to improve energy throughput. As an example[12] of these groups' work, Figure 4 presents (upper curve) the atmospheric absorption spectrum measured from a balloon at an altitude of 40 km on May 14, 1973 during sunrise, showing features due to H2O, CO2 and NO in the 1910 cm⁻¹ region; the lower curve shows a result in the 1890 cm⁻¹ range taken looking at the sun during sunrise from an aircraft flying at 15.5 km with a solar zenith angle of 91.5° on July 13, 1973, again showing H2O, CO2 and NO lines.

Recently an extensive series of measurements by the Girard group have been made from an aircraft during the "Latitude Survey Program"[13]. Data similar to that shown in Figure 4 were recorded at an altitude of about 11 km from 60° N to 60° S, and the spectra were interpreted to yield latitudinal cross sections of the total amount of a number of gases above the flight level. Figure 5 shows such data for NO, NO2 and HNO3[13], three molecules intimately concerned with the stratospheric ozone balance. Clearly a strong latitudinal dependence of HNO3 is seen, though the variation of NO + NO2 is relatively weak.

The results discussed so far have been obtained using incoherent non-monochromatic techniques. A recent result by Mensey[14], employing infrared laser heterodyne methods is remarkable both for the data it produced as much as for the technical advances it represented. In this experiment a laser heterodyne spectrometer[15] operating in the 853 cm⁻¹ region was flown on a high altitude balloon and used to observe the sun along a limb path. The absorption spectra which were recorded as a function of the beat frequency between the incoming radiation and the laser local oscillator is shown in Figure 6. The arrow marks the position of a line due to ClO as measured experimentally: the observed absorption has been interpreted to derive the vertical distribution of ClO which is shown in Figure 7. Also shown in Figure 7 are the results of model calculations of the ClO distribution.[16]

One further area which perhaps merits mention is the development in recent years of cryogenically cooled spectroradiometric devices for IR atmospheric observations. Murcray and co-workers have reported the use of a liquid helium-cooled grating spectrometer[16], and recently a liquid nitrogen-cooled grating spectrometer developed by the NPL group has been flown on two occasions on a high altitude balloon to carry out limb-sounding observations.[17]. The necessity for cooling a spectrometer comes, of course, from the desire to reduce background and stray radiation from and within the spectrometer to a minimum so as to obtain higher radiometric sensitivity to weak atmospheric emissions. Technically, of course, this imposes quite severe difficulties which have, however, been overcome. Measurements in the 8 - 15 um region using such a device[17] have allowed simultaneous observations of the distributions of H2O, O3, HNO3, CH4 and N2O.
At submillimetre or far infrared wavelengths, the situation up to about 1976 has recently been reviewed [17]. At these long infrared wavelengths the atmospheric spectrum is made up primarily of overlapping pure rotation spectra of dipolar atmospheric molecules. Because of this high degree of overlap, high spectral resolution is essential and progress has been defined largely by technical developments which have yielded the possibility of increases in resolution. Since the review given in reference 3, the most significant progress has occurred through the work of two groups.

The first is that at the University of Calgary, which has flown a rapid-scan far infrared Michelson interferometer on balloon flights, producing spectra at a resolution of 0.05 cm⁻¹. These new data, an example of which is shown in Figure 8, have allowed the tentative identification of OH and other spectral features in the very far infrared, for the first time.

The second is that at the Institute for Research on Electromagnetic Waves, in Florence, Italy, which in a collaborative experiment with the NPL has recently flown a very high resolution Michelson (0.004 cm⁻¹) on a balloon: spectral data are still being transformed from the interferograms obtained by this experiment, but first results appear to confirm that the full resolution has been achieved, and that excellent quality spectra should follow shortly.

Solar transmission spectra as measured at a number of tangent heights from a balloon using a laser heterodyne spectrometer. The arrow marks the (measured) position of a ClO line at 853.18104 cm⁻¹. (From Ref. 14)

As an example of the nature of the results which are expected, we show in Figure 9 a spectrum calculated by the NPL group (20), an emission spectrum in the 80 cm⁻¹ region for the case of a balloon (or satellite) effectively above the stratosphere looking towards the atmospheric limb at a tangent height of 30 km. The solid curve shows the principal features of the spectrum, due to H₂O, O₃ and O₂, and the dotted curve shows the effect of adding HCl and HF at a mixing ratio of 10⁻⁹ by volume (constant with height). This calculation clearly indicates that even for mixing ratios 10 to 30 times smaller than the value used, measurable lines due to HCl and HF should be observable.
RECENT RESULTS IN MICROWAVE SPECTROSCOPY

During the past five years or so the determination of upper atmosphere trace gas composition using microwave methods has progressed considerably, primarily through the work of Waters and co-workers. Systems have been developed for ground-based and aircraft-borne measurements of H₂O, O₃ and CO, based on the monochromatic heterodyne methods possible at frequencies below about 400 GHz. A review of microwave atmospheric spectroscopy has recently been presented by Waters.⁴

Figure 10 shows a general view of the atmospheric transmission spectrum from sea level, showing (for a vertical path) the components due to O₂, O₃ and H₂O; O₃ is responsible for the many narrow lines seen between 100 and 300 GHz in the diagram. At the very high spectral resolution (< 1 MHz) achievable at these frequencies (resolving powers of about 10⁵ - 10⁶) it is possible to resolve the "window" regions between the fine structure due to O₃, where weak lines due to other trace constituents, such as CO, N₂O, NO, NO₂, H₂O₂ and others, exist⁴.

As an example of the results which may now be obtained, Figure 11 shows a measurement of the CO line at 115.271 GHz (dots), and the results of calculations of the predicted line shape for three different assumed CO distributions, A, B and C. These profiles are shown in Figure 12: the spectral data of Figure 11 clearly show that profile B provides the best fit to the measurements. This result may perhaps indicate to the reader the high precision for upper atmospheric measurements now possible using microwave systems.

A calculated emission spectrum in the 41 - 42 cm⁻¹ region of the far infrared, for an observer outside the atmosphere viewing the limb at a tangent height of 30 km. The spectral resolution is 0.01 cm⁻¹. The calculation included H₂O, O₂, O₃, HF and HCl, and the significance of the various spectral features is discussed in the text. (From Ref. 20)

6 THE FUTURE

It should be clear from the examples cited above that the spectroscopic study of the upper atmosphere at wavelengths between 1 μm and 1 mm
is a very active field. For the immediate future one can see that certain specific areas are almost certain to show rapid progress: amongst these areas we may include:

- High resolution infrared (3 – 30 μm) and far infrared (30 – 500 μm) spectroscopy using multiplexing Fourier transform spectrometers. The most recent generation of instruments is producing resolutions of 10^{-8} cm^{-1} at 10 μm and 4 x 10^{-3} cm^{-1} at 500 μm.

- Cryogenic spectrometers and interferometers for high radiometric sensitivity and precision for measurements of very weak atmospheric emissions throughout the infrared.

- Laser heterodyne spectroscopy with its capability for very high spectral resolution (eg 10^{-6} cm^{-1}) in the region where many important mid infrared vibration-rotation bands occur between 5 and 75 μm.

- Microwave heterodyne spectroscopy, again capable of very high resolution and also of rapidly improving noise limits as components are improved and as, for example, cooled mixers and detectors become available.

As the variety of available instruments and techniques increases, it is important to remember that different atmospheric investigations demand different instrumental qualities, so that it seems likely that we will continue to require a combination of measurements: it is most unlikely that any one type will provide the panacea for all measurement requirements.

As a final point, we must recognise the growing need to develop satisfactory remote sensing methods in two other areas of geophysical sensing. The first is in the remote determination of upper air winds; to date a number of high resolution methods have been proposed in the visible, infrared and microwave regions, each capable of measuring (with limited accuracy) the Doppler shifts caused by wind motion. This is a difficult measurement problem, however, which still requires a satisfactory solution. The second concerns the remote sensing of the temperature and state of the oceans, which act as huge reservoirs of energy in the atmosphere - ocean system, and which are believed to play a dominant role in controlling the Earth's climate. Here again, difficult measurement problems exist (eg the effect of clouds, haze, molecular transmission, waves, etc) which require detailed study before a reliable remote sensing system can be evolved.

REFERENCES

### TABLE 1
Atmospheric Spectroscopy: A Selection of Groups/Techniques

<table>
<thead>
<tr>
<th>GROUP</th>
<th>COUNTRY</th>
<th>TECHNIQUE</th>
<th>PLATFORM</th>
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<tr>
<td>Stair et al.</td>
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<td>GS/Cryogenic MI/atmos emission</td>
<td>Rocket</td>
</tr>
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<td>Waters</td>
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<td>Zander</td>
<td>Belgium</td>
<td>GS/solar occ</td>
<td>Balloon</td>
</tr>
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Key to Table 1
- IR: Infrared
- Solar Occ: Solar occultation
- GS: Grating spectrometer
- MI: Michelson interferometer
- FIR: Far infrared
- SCR: Selecti: :hopper radiometer
- PMR: Pressure modulator radiometer