Interoffice

NATIONAL RADIO ASTRONOMY OBSERVATORY

TUCSON, ARIZONA

March 2, 1979

25 Meter Millimeter Wave Telescope Memo # 116

To: M. S. Roberts

From: M. A. Gordon

Subject: Absorption of millimeter waves in the atmosphere

1. SUMMARY

A couple of years ago it came to my attention that some research groups had observed a mysterious absorption of millimeter waves in the atmosphere. The observations were made from mountain tops (e. g. Gimmestad et al. 1977). The reports were that the atmospheric transmission over mountains could be twice as large as that predicted from the usual formulas and known amount of precipitable water vapor. The inference is that high mountain sites like Mauna Kea may, on occasion, be less satisfactory for millimeter and submillimeter astronomy than sites at lower altitudes.

Because the British were preparing to check these observations, I elected to wait for their results. Richard Hills, the project scientist for the UK millimeter-wave telescope, has advised me as to their progress. The Science Research Council made a grant to a scientist experienced with Fourier Transform Spectrometers (FTS), Professor D. H. Martin of Queen Mary College, London, to build a state-of-the-art FTS suitable for atmospheric studies. In a recent letter, Hills wrote me that initial measurements with the new FTS failed to show any variable anomalous absorption. One set of measurements were made at 2400 m on Tenerife; another, from the roof at Queen Mary College. It is Hill's opinion that claims for the excess absorption over that predicted by standard atmospheric models are false.

To convince myself, I investigated the phenomenon further by reading a large number of papers on the subject, and by discussing the subject with well-regarded researchers in this field. I now believe that the mysterious absorption exists, thereby disagreeing with Richard Hills. However, I'm uncertain as to the quantitative size of the anomalous absorption and believe it to be a rare phenomenon. Recent observations at sea level also show the anomaly, and I see no reason why it should occur only at high altitude sites. For these reasons, the anomalous absorption may not be important for our Mauna Kea project.

As you know, I have written to Professor Martin, offering to pay travel and expenses for his group to investigate the absorption at Mauna Kea. No answer has been received. Because the anomalous absorption is a controversial subject, I prefer that experts with proper credentials investigate the absorption over Mauna Kea before NRAO enters this research area--time permitting, of course.

2. HISTORICAL BACKGROUND

My impression from reading and the discussions is that the anomalous absorption by atmospheric water vapor consists of a constant and a variable component. The constant part has been known for over 30 years; the variable part, for about 10 years. Only recently has the variable absorption gained credibility. For me it was helpful to consider the historical development of this field, and I shall sketch this below.

The problem of understanding microwave absorption by water vapor has existed for decades. In 1945 Van Vleck and Weisskopf applied their new theory of pressure-broadening to the wings of the atmospheric water line at 22 GHz. In 1947 Van Vleck compared these predictions to the laboratory measurements made by Becker and Autler (1946). He found a large descrepancy. The theory substantially underestimated the observed absorption. The explanations proposed for the anomalous absorption included:

(a) the incorrect modeling of the pressure-broadening, either by theory or by the physical constants of the water molecule,

(b) the unexpected importance of absorption by isotopic species of water vapor, and

(c) the existence of unknown polymers made up of water and oxygen.

Over the years all of these suggestions have been investigated. The problem is still unsolved, but only (a) and (c) are still considered as possible explanations for the anomalous absorption.

Because no simple explanation for the pheomenon was found, interested parties tended to affiliate with one of two groups. The first concentrated on understanding the anomaly in terms of physics and chemistry (H. A. Gebbie is a member of this group). The second, despairing of the slow progress, devised empirical corrections to the radiation transfer equations to permit accurate predictions of the absorption (most radioastronomers belong to this group).

Perhaps the best known of these empiricists are Norm Gaut and Ted Reifenstein. In 1971 they found that the difference between the latest form of the Van Vleck-Weisskopf predictions and the observed water vapor absorption, for frequencies up to 1 THz, could be fitted to within 10% accuracy by an additional correction term proportional to f^2. This correction does not incorporate variations of absorption with temperature, pressure, or time. The existence of the variability was not recognized then, and its dependence on other factors was unknown. For the purposes of telecommunications and radioastronomy, the Gaut-Reifenstein correction is usually adequate. However, it does not explain the anomalous absorption in any way.

The absorption researchers have worked steadily on the problem since 1947. Although the physics (a) and the chemistry (c) suggestions, as well as the absorption itself, have been vigorously investigated, progress has come only recently with the advent of large computers and sensitive radiation detectors. As with all state-of-the-art investigations, the route has had many false paths. The empiricists have had adequate justifications for their approach!

Progress in the physics approach has evidently come to a halt. As Waters (1976) notes in his review article prepared in 1973, the form of the empirical correction gives a clue to the difficulty. The fact that the excess absorption varies as f^2 suggests that the pressure-broadening approximation may not be correct. The absorption coefficient is proportional to the product of f^2 and the Fourier transform of the autocorrelation function of the dipole moment of the water molecule. The f^2 form of the empirical correction means that the Fourier transform must be a constant over a frequency range greater than 1 THz (10^{12} sec^{-1}). Thus, an excessive dipole moment must exist for a time less than 10^{-12} sec, a time comparable to the duration of a collision of atmospheric molecules.

The approximation used to model the pressure-broadening of water lines is that of an instantaneous disruption of the emitted wave train, the so-called <u>impact</u> approximation. The other possible approximation in pressure-broadening theory is the <u>quasi-static</u> approximation, in which the disruption of the wave train occurs gradually over a long period. The typical width of an atmospheric water line in the millimeter region is about 4 GHz at half intensity. If we model the line by a sinx/x function, the Fourier transform is a sinusoidal wave train of duration $(4\ \mathrm{GHz})^{-1} = 2.5 \times 10^{-10}$ sec. This time, the minimum for the water wave train, is 250 times longer than the lifetime of the excess dipole moment, and one is tempted to assume that the impact approximation is adequate. Perhaps not, however. I remember from graduate school days that the impact approximation presumes a phase change of the wave train short compared to the length of a cycle. For the 183-GHz water line, a cycle length is about 5×10^{-12} sec, rather close to the 10^{-12} sec lifetime of the excess dipole moment. Could it be that the impact approximation breaks down in this case?

In other words, the atmospheric situation may be midway between these two standard approximations, a situation currently difficult to handle by known physics techniques. This situation, of course, could account only for the constant part of the anomalous absorption.

The chemists have had more success. It appears likely that under certain conditions of temperature, pressure, and relative humidity that water polymers do form. The simplest of these is the dimer $(H_2O)_2$. In 1967 Viktorova and Zhevakin originally proposed the presence of the dimer to explain the anomalous absorption. Since then, experiments have shown that something is needed in addition to the dimer to account for the excess absorption. Most recently Llewellyn-Jones et al. (1978) have compared laboratory measurements of water vapor absorption with the theoretical absorption by a mixture of water monomers and dimers. They find that at 213 GHz the radiation absorption is still in excess of theory. The amount of excess absorption varied with temperature and with relative humidity (partial pressure of water vapor), suggesting the presence of some additional absorbing process--a nonequilibrium process. The excess absorption increases over the model as a steep inverse function of temperature, it occurs sharply as the temperature decreases below 300 K. It also increases rapidly with the partial pressure of water vapor. This behaviour is what would be expected from the formation of complex water polymers. Here is laboratory confirmation of a variable absorption coefficient, perhaps that seen by Gebbie and his co-workers on Mauna Kea (Moffat et al. 1977).

There are also new results from direct observations of the earth's atmosphere. Since the comparatively crude observations made in 1957 by Gebbie and Burroughs (1968) from Jungfraujoch in the Alps, detector sensitivity has improved considerably (in fact, the combination of high atmospheric absorption and insensitive detectors forced the original investigators to go to mountain tops!). The anomalous absorption has now been seen in the natural atmosphere at sea level, over horizontal paths as long as 500 m (Emery et al. 1979). It is seen to be variable with time, but its statistics are unknown.

3. THE IMPACT ON MILLIMETER- AND SUBMILLIMETER-WAVE ASTRONOMY

Early reports of the variable anomalous absorption may have misled astronomers. As mentioned above, the early observations had to be made from mountain tops for technical reasons. Since then, the phenomenon has also been seen at sea level. I believe that there is no evidence yet that mountains are more subject to the occurrence than sea level sites. However existing observations are quantitatively uncertain, in my judgement, and we may have to wait a long time for a definitive statement concerning the dependence on altitude.

A number of well-regarded observers have looked for the variable part of the anomalous absorption. I tend to agree with Gebbie and his associates that the fact that all of them do not see the effect indicates that the anomaly is truly time-variable. The laboratory work of Llewellyn-Jones et al. (1978) tends to confirm this.

Hans Liebe (of whatever NBS Boulder is now called), a respected researcher in this area, emphasizes that many highly regarded scientists agree that the variable effect exists and, further, that it should exist on theoretical grounds. The formation process of the water polymers is now being referred to as "ion-clustering", a process by which atmospheric radicals combine to form complex polymers. Liebe, who is privy to his colleagues unpublished opinions, told me that there is enough of a consensus among atmospheric scientists as to the problems of the water vapor absorption that there will be a special conference this September in Vail, Colorado, to discuss the subject. He feels that the quantitative aspects of the pheonomenon seen over mountain tops are fairly uncertain because of observational difficulties, compared to the carefully prepared laboratory and sea level work.

In conclusion, I don't know how much of a problem the variable anomalous absorption is for our planned operation at Mauna Kea. It could be very weak when it does occur. It could occur extremely infrequently, and then only during the afternoon, when convection brings water vapor to the summit area. To investigate these aspects is a challenging project, and certainly one which will require a large number of days on Mauna Kea. From what I now know about this problem, I'm reluctant to suggest that the NRAO become involved in the experimental investigation of the variability of radiative absorption by atmospheric water vapor. It's a knotty problem.

For background information, I'm attaching a plot of the atmospheric transmission at Kitt Peak.

c: R. E. Hills
D. E. Hogg
25-m Telescope Working Group
B. E. Turner
J. W. Waters

TEXT REFERENCES

Becker, G. E. and Autler, S. H. 1946, Phys. Rev. **70**,300.

Emery, R. J., Zavody, A. M. and Gebbie, H. A. 1979, *J.A.T.P.* (in press)

Gaut, N. E. and Reifenstein III, E. C. 1971, Environmental Res. and Tech. Report 13, Lexington, Massachusetts.

Gebbie, H. A. and Burroughs, W. J. 1968, *Nature* **217**, 1241.

Gimmestad, G. G., Ware, R. H., Bohlander, R. A. and Gebbie, H. A. 1977, *Ap.J.* **218**, 311.

Llewellyn-Jones, D. T. Knight, R. J., and Gebbie, H. A. 1978, *Nature* **274**, 876.

Moffat, P. H., Bohlander, R. A., Macrae, W. R., and Gebbie, H. A. 1977, *Nature* **269**, 676.

Van Vleck, J. H. and Weisskopf, V. F. 1945, *Rev. Mod. Phys.* **17**, 227.

Van Vleck, J. H. 1947, *Phys. Rev.* **71**, 425.

Viktorova, A. A. and Shevakin, S. A. 1967, *Sov. Phys. - Dokl.* **11**, 1065.

Waters, J. W. 1976, in Methods of Experimental Physics **12B**, 142-176, (edited by M. L. M eks, Academic Press, New York).

OBSERVATION REFERENCES

Jungfraujoch: Gebbie and Burroughs (1968)
Gebbie, H. A., Chamberlain, J. and Burroughs, W. J. 1968, *Nature* **220**, 893.

Baja Observatory: Gimmestad, G. G., Gebbie, H. A., Bohlander, R. A. and Mendoza V, E. E. 1972, *Ap.J.* **178**, 267.

Mt. Evans: Gimmestad *et al*. (1977).

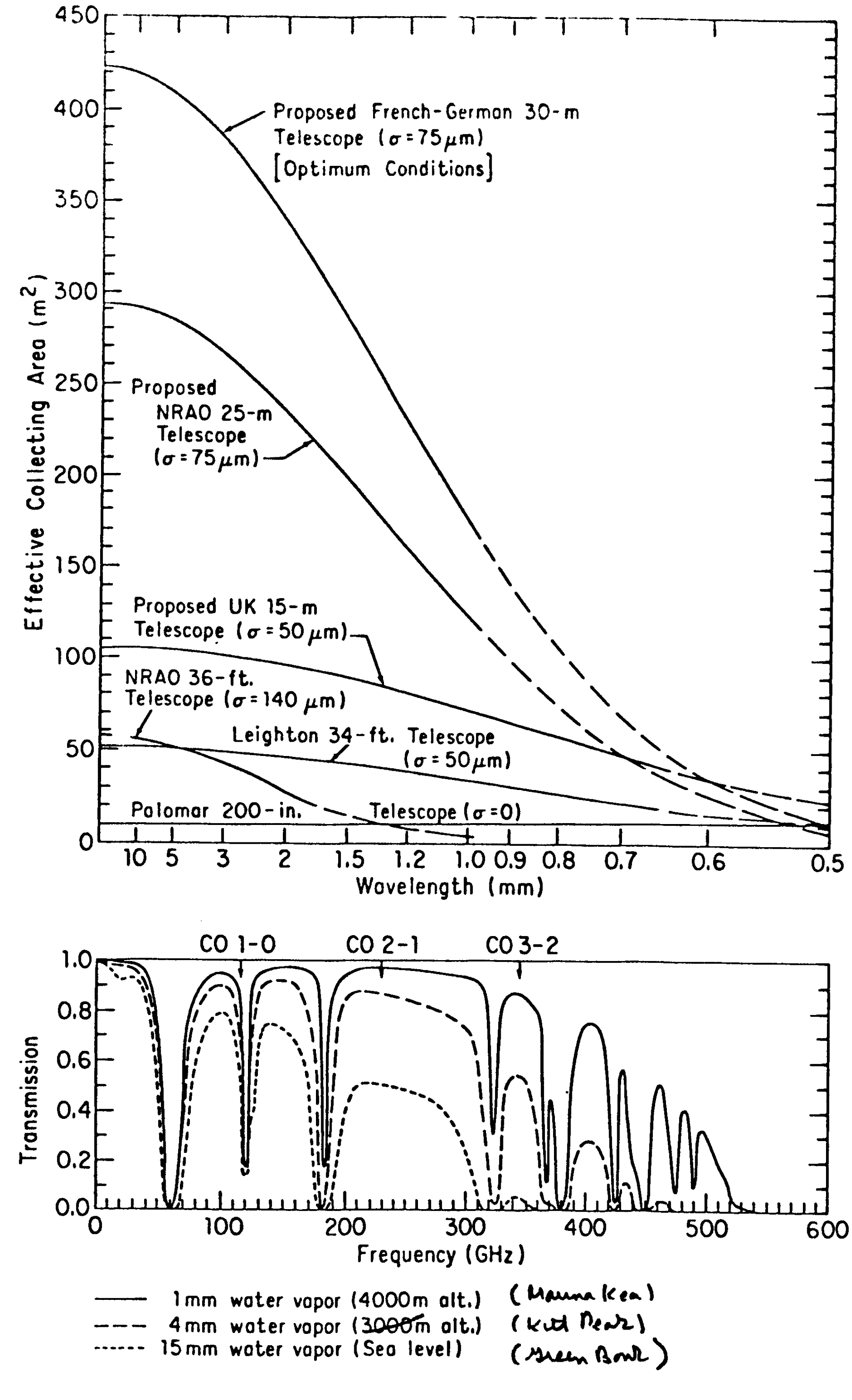
Mauna Kea: Moffat *et al*. (1977)
Nolt, I. G., Martin, T. Z., Wood, C. W. and Sinton, W. M. 1971, *J.A.T.P.* **28**, 238.

White Mountain: Mather, J. C., Werner, M. W. and Richards, P. L. 1971, *Ap.J.* **170**, L59.

Tenerife: Hills, R. E., Webster, A. S., Alston, D. A., Morse, P. L. R., Zammit, C. C., Martin, D. H., Rice, D. P. and Robson, E. I. 1979 (preprint).

Laboratory: Llewellyn-Jones *et al*. (1978).

Sea Level: Emery *et al*. (1979).



Mort: Here is a plot comparing the "constant" absorption (transmission) on Mauna Kea with the telescope performance

Figure 4. Top: The variation of collecting area as a function of wavelength for a few telescopes useful at millimeter waves. Bottom: The transmission of the atmosphere at 3 levels of precipitable water vapor (from the SRC proposal).