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A STUDY OF WATER AEROSOL ABSORPTION AND EMISSION EFFECTS (INCLUDING FOGS) IN THE 8- TO 13- MICRON INFRARED WINDOW

Hugh R. Carlon

Edgewood Arsenal
Edgewood Arsenal, Maryland

December 1969

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DEPARTMENT OF THE ARMY
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FOREWORD

The work described in this report was conducted under Task 1B662706A10202, Area Scanning and Warning Techniques. This work was started in January 1969 and completed in October 1969.

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DIGEST

Water aerosols theoretically are capable of very strong absorption and emission in the 8μ to 13μ infrared atmospheric window, owing to the 10^4 increase in the absorptivity of water in this spectral region for the liquid phase as compared to that for water vapor. The author reviews his earlier papers in light of subsequent developments reported in the literature and finds strong evidence for water aerosol activity in measurements relating to atmospheric transmission, radiance, and turbulence. Several examples are given of experimental increases in optical activity of the atmosphere with relative humidity increases, even though absolute humidities (reflecting water vapor concentrations normally stated to cause similar effects owing to wing absorption of distant lines) are quite constant. Availability and suitability of condensation nuclei are thought to be significant in determining the degree of water aerosol absorption and emission. Emission effects are *most pronounced*.

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A STUDY OF WATER AEROSOL ABSORPTION AND EMISSION EFFECTS (INCLUDING FOGS) IN THE 8- TO 13-MICRON INFRARED WINDOW

I. INTRODUCTION AND REVIEW OF PREVIOUS WORK.

In a previous journal article,[1] the author presented a theoretical development explaining how water aerosols in equilibrium with water vapor at higher relative humidities could cause significant attenuations of infrared energy in the 8μ to 13μ infrared window, provided that sufficient condensation nuclei were available. This effect originates in the differences between the spectral absorption, or "extinction," of equal quantities of water vapor versus the liquid phase, as an increase in optical density of approximately four orders of magnitude (10^4) is observed in liquid water over that for the vapor phase. Originally, this theory was developed to explain observed changes in terrestrial scintillation (shimmer) intensity with changes in humidity, but it soon became apparent that other optical phenomena involving the atmosphere might have explanations implicit in this work. A second paper[2] explained how cool, humid, invisible air masses at higher altitudes might contribute to lowered satellite measurements of the surface temperature of the earth under apparently clear conditions, owing to absorption and/or emission effects of these damp air masses. More recently, Hall[3] pursued this approach in making a study of the specialized case of ice crystal (cirrus cloud) contributing to infrared sky radiance. A further extension of the theory led to a possible scheme for the detection of clear-air turbulence (CAT) ahead of aircraft, since CAT is frequently accompanied by humidity gradients. This work was also well-documented in Edgewood Arsenal publications,[4-7] as much of the effort resulted from problems associated with the E49 LOPAIR (Long-Path Infrared) toxic agent alarm program.[8] Notably, these were problems of atmospheric scintillations leading to instrument instability and variations in instrument signal levels and optical balance under varying meteorological conditions. For this reason, fog transmisson studies, performed recently by the author, utilized modified E49 equipment. These studies will be discussed presently. A continuing in-house program is directed toward the study of related atmospheric effects, as these have a direct bearing upon the potential performance of many candidate agent detection systems now in feasibility study or in exploratory development in these Laboratories. These include laser devices, passive LOPAIR hardware, and most especially, nonspecific LIDAIR (laser-radar) systems.

II. A RE-EVALUATION OF THE PREVIOUS WORK.

Since the publication of the original paper,[1] relating to the water aerosol theory, the literature has been searched at length to locate data which would be useful in evaluating this theory. At least four good references were located describing liquid water absorption in the 8μ to 13μ region.[9-12] Of these, the work of Centeno[12] is often cited, and his values of absorption were used in place of those mentioned in the original paper, which were run in-house, for purposes of computation. Centeno also developed a dispersion curve for liquid water in the spectral region of interest. This curve is of considerable value in our discussion, and is presented as figure A-1.* Wyatt, Stull, and Plass,[13] published comprehensive absorption values for water in the vapor phase. Curves developed from their data for 1050, 1100, and 1150 cm^{-1} are presented in figure A-2 in terms of centimeters of precipitable water versus percent optical transmission. Calculations of the ratio of absorption of water in the liquid phase to that in the vapor phase were made (shown in table I of our original paper) and values assigned to the ratio of between 10^3 and 10^4. When the more precise absorption figures (from the literature just

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* Figures A-1 through A-13 are in appendix A.

discussed) are used to compute this ratio in the vicinity of the 10μ wavelength, it is seen that this value is even somewhat larger than was originally estimated, and is greater than 10^4 in almost all cases. That is, in condensing from vapor to liquid phase, water becomes even more opaque in the 8μ to 13μ region than had been postulated previously as the basis for the large optical attenuations theoretically possible under conditions of high humidity. If we neglect, for the moment, scattering contributions from water aerosols (assuming very small droplets with respect to the wavelengths of interest at 8μ to 13μ), it is possible to compute the theoretical transmission over various optical paths as a function of air temperature and resulting humidity content. This is done in figure A-3. These curves are based upon the saturation water content for the temperature scale shown, and are computed from the data of Centeno. In effect, these curves show the computed transmission for saturated air at various temperatures which would exist if it were possible to suspend the atmospheric water entirely as true liquid droplets so small that 10μ scattering were negligible. In practice, fortunately, it is seldom possible to meet the criterion that all water in an atmospheric volume be suspended as true liquid in small droplets. In nature, this virtually never happens. In the laboratory, it is possible to generate small droplets artificially by boiling, as discussed in the experiments of the original paper.[1] When such drops are generated, the result can be striking – the visible wavelengths may be much less severely attenuated than are those in the 8μ to 13μ window. In nature, condensation of droplets at higher relative humidities depends upon the availability and suitability of condensation nuclei. Rarely are these present in sufficient numbers to provide the amount of droplet cross-sectional area necessary for dramatic infrared signal absorption. Rather, the effect would be typically a troublesome one, accounting for anomalies of perhaps 10% or 15% in transmission readings (although frequently more in emission readings), often going undiscovered or misunderstood among other sources of potential error such as instrument noise. In naturally-occurring aerosols, a wide droplet size distribution is commonly present and Mie scattering also becomes an important factor in the determination of optical transmission through the atmosphere, as discussed in the original paper. However, figure A-1 indicates that the magnitude of Mie scattering in the 11.5μ region* may be much less than was previously supposed, owing to the anomalous dip in index to about 1.11, which may be attributed to the absorption of liquid water in this vicinity. In that case, pure absorption of the droplets would be proportionately more significant.

We see, therefore, that the spectral concepts set forth in the original paper[1] are valid and that the magnitude of predicted theoretical effects is greater than previously estimated. But while figure A-3 would show a potential attenuation of 50% through only 1 meter of saturated air at a temperature of less than 50°F., we know in practice that this simply does not occur, even in the case of fogs where scattering is also an important contributor. The resolution of this dilemma must, therefore, lie in the availability (size, suitability, and numbers) of condensation nuclei found in nature. The potential magnitude of this effect must, however, be appreciated.

III. RELATED LITERATURE AND EXPERIMENTAL FINDINGS.

The author, since the publication of his original paper,[1] has conducted an extensive literature search and has collected a large number of references which are directly related to the water aerosol theory. Sabbagh,** in a private communication, concurred with our emphasis upon the significance of absorption and emission by water vapor and droplets in the 10μ region by stating his conviction that measurements of the emissions by these absorbers generally can be of far-reaching importance to problems in meteorology. He cited the work of

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* A commonly-used wavelength for satellite measurements is 11.1μ.

** Sabbagh, E. N. Senior Meteorologist, Singco, Inc. Burlington, Massachusetts. Private communication. February 1967.

Oetgen, Bell, Young, and Eisner[14] in spectral radiance measurements of the sky at wavelengths between 1μ and 20μ, where the authors observed considerable activity in emission in the 8μ to 13μ region: this they attributed to the wings of water vapor absorption bands (a commonly-accepted alternate explanation for such activity, although difficult to justify when large variations are noted, as is often the case). These authors further noted that additional water content in certain of their tests, as opposed to others in drier climates, gave rise to greater emissivity in the 8μ to 13μ wavelength region and almost unit emissivity at wavelengths greater than 15μ (where CO_2 also contributes). Goody[15] studied this region using a high-resolution spectrometer, and he noted that a co-worker had investigated atmospheric absorption and emission in the 10μ region, concluding that there is probably a large background contribution from *nongaseous constituents.* This will be discussed in a later section. Clark[16] recently described an observed effect similar to that postulated in our original papers. In measuring the temperature of the sea with an airborne infrared radiometer, he experienced errors of up to several degrees centigrade owing to what he calls "nascent clouds" lying above the water's surface. He described these as developing eventually into fog patches, and initially covering up to several hundred square miles, but since they are *invisible*, they are capable of eluding the observer's ken. Nordberg,[17] reporting to a specialty group on TIROS, in 1964, noted that difficulties had arisen in estimating surface temperatures in apparently clear air, that the readings in one case were consistently low, and that such factors as surface emissivity were considered to adjust results. At the same meeting, Blau's observation that some water haze existed, even in clear air, was discussed, and it was further stated that he had noticed spectral differences between ice and water-drop reflections (a phase transition). The literature contains many references such as Wexler's,[18] discussion of low surface temperature readings obtained at 8μ to 13μ by TIROS III and other satellite and balloon flights. We shall see that the *potential* effects of water aerosol emissions are enormously important in considering the design and operation of any *passive* infrared radiometer.

In the area of atmospheric transmission, data such as those of Taylor and Yates[19] have become standard reference works. Streete,[20] in his measurements of atmospheric transmission at sea level, over a 25-km path, was unable to obtain agreement between his measurements near 10μ wavelength and standard reference curves, even though approximately the same amount of water vapor was present for each of the curves examined. He noted a considerably higher transmission at 56% RH than appeared in the reference spectrum taken at 82% RH, and he further observed that relative humidity was the only significant difference in meteorological conditions under which the results were obtained. He cited our paper[1] as a possible explanation. Burch[21] described measurements of water absorption in a 30-meter cell, noting a considerable attenuation in the 8μ to 12μ region which he attributed to the extreme wings of lines centered on either side of this region. He described the difficulty encountered in making these measurements, and he observed that the usual band models employed in other spectral regions were not applicable here. Water, even at a partial pressure 0.01 that of the inert cell atmosphere, accounts for most of the absorption in this spectral range. Burch related this attenuation mathematically to the square of the water pressure.

Our original paper addressed the problem of terrestrial scintillation directly, and it would be a mistake not to discuss the more recent papers which take note of the apparent relationship of this phenomenon, or of similar ones, to atmospheric humidity levels. The author, in a recently concluded series of transmission measurements, discussed below, appreciated for the first time how readily increasing humidity must contribute to scintillation, at least in the visible wavelengths, since the formation and growth of haze under these conditions, with its attendant Rayleigh scattering, must cause optical beam modulation in the presence of any convective air movement. Makarova, et al[22] described summer conditions at a mountain test site where a water variation in the atmosphere of 2:1 was observed from morning to noon as the result of melting snow. This led to instability of the optical properties of the atmosphere. Alcott[23] summarized work performed by the British Aircraft Corporation

(BAC) which showed that inhomogeneities in developing fog may cause effects similar to turbulence-generated scintillation. In this study, Newton, Lavin, Titmuss, and Williams[24] examined atmospheric transmission over a horizontal range 3 feet above the ground, using a cine recording system. A gallium arsenide laser with suitable optics was used as a source. The researchers noted a different type of optical activity associated with developing fogs which could not strictly be called a turbulence effect since it did not seriously degrade the projected pattern, but which, nevertheless, produced very high modulation values with little blurring of the projected edge, although this edge did show discontinuities. Figure 4 presents an illustration from their paper, showing three motion picture film takes corresponding to neutral, developing fog, and normal turbulence conditions, respectively. These results, which were taken at a range of 3000 yards, are stated to clearly establish *the existence of a mechanism peculiar to fog conditions.* Newton and his co-workers concluded that this work has shown that there is a new mechanism causing modulation that is associated with the development of fog. Our water aerosol theory offers a ready-made explanation of this mechanism.

In a remarkable paper, Nichols and Lamar[25] reported results using an instrument which converts infrared images into visible color images in which the colors are representative of different infrared wavelength regions. These wavelength regions, detectors used, and conversion colors in the visible are as follows: 0.5μ to 1.0μ, Si, blue; 3.0μ to 5.5μ, InSb, green; 8.0μ to 14.0μ, HgGe, red. Figure A-5(a) shows a photograph taken across a desert floor on a warm and somewhat humid day. Figure A-5(b) shows the same scene as observed in the 8μ to 14μ band and reproduced in red on the conversion image. The authors explained that red appears when cool, but highly emissive, objects are in the scene. On warm and *humid* days, they stated that the atmosphere itself can be seen glowing incandescently, as shown in the illustration. Furthermore, it is noted that the 8μ to 14μ atmospheric window is relatively transparent, but self-emission of the air occurs near the horizon and where sufficient warm water vapor is present. On clear, dry days this self-emission is not as evident, and pictures with horizons 40 to 50 km away stand out in sharp contrast. This is, of course, a matter of degree and is subject to gain settings as well as atmospheric effects.

Nichols* noted that it is usually possible to equate good seeing in the visible to good seeing in the 8μ to 13μ window, but that this is not always so, as shown in figure A-5. While he has observed poor visibility with a relatively good 8μ to 13μ window, a common occurrence, and one often noted when looking into fog banks, for example, he has also encountered examples of *good visibility with a poor 8μ to 13μ window,* the sort of thing which we would expect from the water aerosol theory under discussion. Nichols stated that the whole situation depends largely on the amount and the state of the water in the air. He noted, as in the paper already cited,[25] that the 8μ to 13μ window is subject to increasing emission as transmission drops (absorption increases), blocking clear sight of the object being looked at in the distance. Fortunately, in his desert environment, these problems do not occur too frequently.

IV. MAGNITUDE OF WATER AEROSOL EFFECTS.

We stated previously that the magnitude of optical effects which we would expect to be attributable to water aerosols, given sufficiently high specific and relative humidities, should be dependent to a great extent upon the availability, size, and suitability of atmospheric condensation nuclei. Kruse, McGlauchlin, and McQuistan[26] presented a discussion of this

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* Nichols, L. W. Naval Weapons Center, China Lake, California. Private communication. October 1968.

availability for clean country air, industrial-area air, and also for clouds and fogs. We shall discuss each of these in turn.

Clean country air, under conditions of low humidity and very high visibility, contains approximately 10^5 particles per liter. About 95% of these are of radii 0.1μ to 1.0μ, with the balance 1μ to 10μ. From the curves in our original paper,[1] we see that even if it were possible to condense into droplets all water available in saturated air containing this concentration of nuclei, the resulting cross-sectional area would be only about 10^3 mm^2 per liter or less. Furthermore, for all moisture to be accommodated, the drops would have to average more than 10μ in radius. This would lead to Mie scattering in the 8μ to 13μ wavelength region. Clearly, in this case, the water aerosol absorption effect would be virtually nonexistent. To begin with, the relative humidity under actual noted conditions would be too low to stimulate droplet growth. That there is virtually no growth is indicated by the very high visibility cited, indicating very little Rayleigh scattering even at visible wavelengths.

The pale blue haze hanging over industrial areas, which may rise several thousand feet, is composed of particles of radii 0.03μ to 0.2μ. Rayleigh scattering in the visible is, of course, very pronounced. Approximately 10^8 particles are found per liter of air. These could give a cross-sectional area of less than 10^4 mm^2 if it were possible to condense all water in saturated air entirely upon their surfaces. This would still result in a required average droplet radius of about 1μ or more, which would enter the Mie region for 8μ to 13μ wavelengths, making it difficult to tell true aerosol absorption from scattering in transmission, although the absorption contribution could still be observed by studying *emission* from the atmospheric sample. Furthermore, it is clear that all atmospheric water could not be absorbed onto these nuclei, that all drops would not be of similar size, and that droplet growth is still dependent upon relative humidity levels. For these reasons, it is considered highly unlikely that more than, say, 1% of atmospheric water would become effective in the type of attenuation contribution which we are discussing, except in unusual meteorological circumstances. Thus we see that water aerosol absorption effects in the 8μ to 13μ window, while rarely overwhelming, could be expected to play a significant role in the determination of atmospheric transmission values in the presence of suitable nuclei. These effects probably help to account for some of the variances in transmission, as reported in the literature and discussed in the last section. Also, it should be remembered that water aerosol, owing to anomalous effects already discussed and illustrated in figure 1, may act as an almost pure absorber in the 11.5μ wavelength region where its refractive index drops to 1.11 and Mie scattering is consequently diminished. From the standpoint of emission, as affecting the performance of systems such as passive infrared radiometers, and as reported in this spectral region by Nichols[25], * *we may expect this water aerosol effect to be much more pronounced.*

Clouds are composed of droplets of radii 2μ to 30μ, and they typically contain between 5 X 10^4 and 1.5 X 10^6 droplets per liter of air. This concentration range yields cross-sectional areas of from 10^2 to 10^3 mm^2 per liter and a droplet radius distribution, as already noted. For the 8μ to 13μ window, these droplets are squarely in the Mie scattering region, and scatter and droplet absorption would be expected to take on approximately a 1:1 relationship. From our theory, a cloud at 27°C would be expected to give a 50% attenuation of a transmitted beam through about 0.3 meters of its main body. Understandably, such estimates are difficult to verify experimentally, even though some measurements of cloud transmission have already been performed capably.[27] The main difficulty lies in determining the path lengths accurately, as well as the homogeneity of the cloud mass. Larger cloud droplets, in contradiction of Mie calculation assumptions, are essentially "black."

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* Nichols, L. W. Naval Weapons Center, China Lake, California. Private communication. October 1968.

Fogs are composed of droplets having radii in the 3μ to 60μ range. Typically, 10^3 to 5 X 10^4 drops per liter are observed. Wolfe[28] gave a good review in this area, and the work of Arnulf and Bricard[29] is classical. If all fog droplets become well developed or "aged," they will present a cross-sectional area of from 10^2 to 3 X 10^2 mm^2 per liter, exhibiting Mie or nonselective behavior at 8μ to 13μ wavelengths and clearly nonselective behavior for visible light. The water aerosol theory, under discussion, would predict an attenuation of perhaps 50% through 5 meters of fog and a 10μ wavelength, although this attenuation falls rapidly for larger droplet sizes. This figure corresponds to an optical density of 60, which is very high but is within a factor of 3 of some observed points reported for this wavelength by Arnulf and Bricard. Johnston and Burch[30] reported fog attenuations at 10μ, approximately one-half those in the visible, and these results agree well with other data in the literature, as well as with the author's measurements, which will be discussed shortly. Rensch[31] made measurements of laser attenuation through fogs at 0.6328μ and 10.59μ wavelengths. Transmission studies do not, of course, explore emissive behavior of fogs in the 8μ to 13μ window.

V. LOPAIR FOG TRANSMISSION STUDIES.

We have seen that transmission studies are not ideal as a tool in studying water aerosol effects; rather, emission studies would be more productive and are, in fact, under way using equipment described below. Nevertheless, since water aerosol absorption can be expected to have some bearing upon atmospheric transmission, particularly in the case of clouds or fogs, it was considered worthwhile to make a series of transmission measurements using modified E49 LOPAIR[8] equipment. Two E49 LOPAIR transceivers were set up on a platform overlooking a wooded ravine and a stream in an area frequented by fogs. The transceivers were placed several feet apart to eliminate "crosstalk" between the instruments. One transceiver was modified for operation in the visible, using a CdS detector peaked at 0.515μ and glass optics. The Nernst glower source was used since power supply circuitry was already built into the head. The second transceiver was left unmodified, using germanium optics and an immersed thermistor bolometer detector. A mosaic of 33 cube-corner reflector elements was set up on the opposite bank of the stream, 150 feet from the transceivers. This permitted a folding of the optical paths to give total path lengths of 300 feet (.0914 km). The systems ran continuously through the fall and winter months of 1968 and 1969, except for down-time necessitated by repairs or general maintenance. A sling psychrometer was used to make temperature and humidity measurements at the same time that preamplifier RMS output voltages were monitored from both transceivers. The spectral response of the unmodified system was adjusted to give a broad-band sensitivity between 9μ and 12μ wavelengths, as shown in figure 6. This was easily accomplished, since the LOPAIR equipment utilizes "natural-type" plastic optical filters[32] which may be tailored for spectral response. This broad infrared response was used to integrate absorption measurements through most of the window, making the system insensitive to small, spurious absorptions. A spectral scan performed through 100 feet of fog had shown little specific spectral activity in this wavelength region (figure A-7). This scan was taken using a specially-designed and highly versatile spectrally-scanning radiometer, built to Defense Development and Engineering Laboratories specifications by Exotech, Inc., Rockville, Md. This instrument, using a circular-variable filter, can spectrally scan the region from 2μ to 15μ over a range of scanning rates and time constants, using either a cooled or bolometer detector, operating either actively (remote infrared source) or passively (a mode which is very useful in atmospheric emission studies). The optics can be focused from a few feet to infinity, and readout is by tape, strip chart recorder, or oscilloscope/photograph combination. Figure A-7 was normalized to the blackbody curve for clear, sunny conditions.

These fog transmission spectra are extremely useful, and they will be gathered and reported as circumstances permit.

Figure A-8 shows the relationship between optical density, as used by Arnulf and Bricard,[29] and percent signal transmission for the experimental conditions of the LOPAIR fog trials. Arnulf and Bricard noted densities at the 10μ wavelength ranging up to 20 km^{-1}. This would correspond to a transmission of about 1% in the LOPAIR trials, as may be seen from the curve.

The relationship between attenuation in the visible and the 9μ to 12μ wavelength regions for a typical fog is shown in figure A-9. This fog, which occurred on 13-14 October 1968 at an air temperature of 61°F, was remarkably consistent in that points taken during both development and dissipation of the fog tended to follow the same curve. The 45° line is included simply to show the deviation from the 1:1 attenuation actually obtained. The 2:1 attenuation ratio of visible to infrared (10μ) wavelengths, noted by Johnston and Burch,[30] is approximated over a wide range of our observed attenuations. Under extremely heavy fog conditions, which might popularly be characterized by the term "pea soup," infrared transmissions of about only 6% were noted, corresponding to an optical density of 13 (3% and optical density = 16.5 in the visible). Figure A-10 shows the curve for a very fine mist which eventually developed into a fine fog, but cleared before long-aging could take place to appreciably increase droplet size. The point lying close to the y-axis is interesting here. For the two wavelength regions, it can be seen that the condensation of liquid water on the mosaic reflector surfaces immediately results in a shifting of the ratio of energy returns to the opposite side of the diagonal. In this case, an increase in absorption of about 30 times has taken place over that which would be typical for the airborne aerosol at the 9μ to 12μ wavelength region. Shortly after this curve was obtained, the mosaic cleared in sunlight and the transmission level went to 100%. Figure A-11 shows data for a long-lasting fog on the evening of 31 December 1968 to 1 January 1969. Owing to the stability of this fog, very few points could be obtained except in the area shown. The five points presented were taken through a 5-hour period.

The fog data presented here have been corrected for such factors as CdS detector sensitivity drift with temperature and background noise level for both transceivers. Comparative measurements were much easier to make during hours of darkness, since in daylight large and varying amounts of reflected background light reached the visible detector.

While these data presented no significant departure from previously reported measurements, and observed results were about as expected, the observations indicated that water absorption probably does make a contribution in the 8μ to 13μ window region, certainly in the case of condensation of even trace amounts of liquid water on the optics of active infrared equipment operating in this range. It was also noted that, in the location where these measurements were made, the availability of condensation nuclei was not sufficient to cause appreciable attenuation of infrared signals with increasing relative humidities, short of actual fog formation.

VI. CONCLUSIONS.

Water aerosols theoretically are capable of very strong absorption and emission in the 8μ to 13μ infrared atmospheric window, owing to the 10^4 increase in the absorptivity of water in this spectral region for the liquid phase as compared to that for water vapor. The author reviews his earlier papers in light of subsequent developments reported in the literature, and finds strong evidence for water aerosol activity in measurements relating to atmospheric transmission, radiance, and turbulence. Several examples are given of experimental increases in optical activity of the atmosphere with relative humidity increases, even though absolute humidities (reflecting water vapor concentrations normally stated to cause similar effects owing to wing absorption of distant lines) are quite constant. Availability and suitability of condensation nuclei are thought to be significant in determining the degree of water aerosol absorption and emission. Emission effects are *most pronounced*.

VII. GUIDELINES FOR FUTURE INVESTIGATIONS.

If there is one aspect of water aerosol droplet growth theory which is most important and least understood, it is probably that point in the condensation of water about a nucleus at which the water ceases to be vapor-like and becomes liquid-like. Typical condensation nuclei have radii of about 0.03μ to 0.1μ. These alone often cause haze owing to Rayleigh scatter in the visible wavelengths. As relative humidity increases, water molecules begin to adsorb on these nuclei. The molecular radius of water vapor is approximately $10^{-4}\mu$, so that a great many water molecules may attach themselves initially to the comparatively vast nucleus without coming together. Eventually, growth of the droplet becomes discernible, indicating that water, many molecular layers thick, had built up on the surface of the nucleus. Even before the drops are large enough to significantly scatter infrared energy in the 8μ to 13μ wavelength region, they begin to absorb significantly (assuming that the water in these droplets is now behaving like a liquid). By the time the droplets are Mie scattering at 10μ, they are already capable of absorbing about 50% of a diametric ray. If the drops become large enough and if they have formed about a particularly soluble nucleus, they may dissolve the nucleus altogether, perhaps adding spectral qualities of the solute to the picture. Given an ambient air temperature, these absorbing droplets will emit in the 8μ to 13μ window region. It is this emission which is considered to be of importance as a subject for further study.

Appendixes B and C contain two short technical papers discussing further implications of our water aerosol theory beyond those discussed in the open literature.[1,2] Both of these papers are unpublished, and are presented here for the first time. The first deals with the possibility of CAT detection ahead of aircraft using modified radiometric equipment similar to that now commercially available, while the second deals in considerable detail with the interpretation of satellite measurements of earth surface temperature using the water aerosol explanation of temperature deviations from true values.

At least three detailed papers have been published dealing specifically with atmospheric emission or radiance in the 8μ to 14μ wavelength interval.[33-35] Roach and Goody,[33] in reporting the results of work[15] mentioned earlier, presented a hypothetical explanation, beyond that readily explainable by selective absorption of atmospheric gases, of their observation of continuous extinction recorded over Ascot and London, England. This continuum showed increased opacity toward longer wavelengths. The opacity was much greater in London than in Ascot, where the air was relatively clean, and although it correlated with water vapor content at both locations, the authors were forced to conclude that *aerosol extinction* accounted for observed data. This sounds reasonable in consideration of our earlier discussion of condensation nuclei, which would certainly be plentiful over London. It was, in fact, in an attempt to resolve whether or not this continuum exists, and if so what its cause may be, that Roach and Goody undertook this work. They developed the two logical alternatives to explain their observations, namely, gaseous extinction or particulate solid or liquid. If gaseous, this must arise from the residual effect of the far wings of distant strong lines, as described by Elsasser.[38] in 1938. If particulate, the authors consider spherical water droplets or irregular solid particles. Earlier work in Germany had shown that dry particulate aerosols could give extinction coefficients of only about 2×10^{-3} km^{-1} near 10μ wavelength, 50 times too small to account for the results obtained. Therefore, water droplets were considered in detail, and although interpretation of the experimental results was not conclusive, the potential effects of these aerosols were noted as probably contributing significantly to the data obtained. In a sense, this paper anticipated the discussion developed in our present paper. It was not possible for Roach and Goody to draw firm conclusions, however, owing to lack of available information concerning droplet concentrations under the "time and place" conditions of these experiments. When we compare the experimental extinction coefficients of Roach and Goody's (figure A-4) to these coefficients for *liquid* water.[9-12] the similarity in the 8μ to 13μ region is most striking. It is interesting to note that this paper references the work of H. L. Wright in the growth of water droplets about condensation nuclei, from the standpoint of optical scattering of the droplets thus formed. It should be noted here that Mr. Wright, still active in England at the time of preparation of our original paper.[1] was sent a copy of the manuscript describing water absorption, as opposed to scattering, by these droplets. He found the idea quite plausible and was interested in the water absorption theory to the point of making several helpful additions to the manuscript.

In a more recent paper, Griggs[36] reported measured and calculated earth-atmosphere radiance data in the 8μ to 14μ wavelength region as a function of altitude. Results are presented in a series of graphs showing good emission levels in this window region, particularly for warm, moist atmospheres where radiance levels perhaps four times larger than those for cold, dry atmospheres are noted. Some disagreement is cited in the 7μ to 9μ wavelength region which is "not readily explained". It is thought that the discrepancy is probably due to different continuum absorption models used. In the 11μ to 13μ region, agreement is good for the moist target and poor for the dry one. An interesting observation is made that, when sighting on clouds, care must be taken to avoid seeing the earth's surface below, which is usually warmer than the cloud. This is the converse of the surface measurement problem which we discussed earlier,[2] where one wishes to exclude nascent clouds from being in his field of view without his knowledge as he attempts to determine surface temperature. Griggs concluded with a highly pertinent discussion of measurements of the temperature of the earth's surface from an altitude of 5.49 km, stating that *readings are as much as 4°C lower than actual surface temperature*. Our earlier paper[2] would have predicted precisely this behavior in the presence of nascent clouds. Further, Griggs continued, when the atmosphere is much *warmer* than the surface, the measurement can be *higher*, again, exactly as would have been predicted by our theory. In this case, a 2.2°C error was noted. He stated further that the atmospheric effect is not always negligible even at the lowest altitudes, as in an observation where the surface temperature read 0.3°C high at an altitude of only 0.05 km, and 0.9°C high at 0.15 km, presumably under inversion conditions.

An earlier paper[37] also reported striking variations in atmospheric emission, with temperature and humidity, far larger than could be explained by vapor concentrations alone. The formation of water aerosols at higher relative humidities would seem a much more logical explanation.

To the present time, the contribution of water droplet absorption in the 8μ to 13μ window region, as opposed to vapor absorption or water droplet scattering, has not been resolved. There has been, in fact, a tendency to ignore this contribution as a troublesome encumbrance of existing theories and atmospheric models, even though the models frequently fail to fit observed data in this wavelength region. The effects of droplet absorption and emission are frequently observed (or so it seems, in work cited in these pages), and yet are very difficult to measure experimentally owing to the complicating factors of relative humidity, absolute humidity, availability and suitability of condensation nuclei, and numerous less significant parameters. Perhaps a satisfactory model which would include these aerosol effects simply would be too complex to put into practical use, or for that matter, too complex to develop in the first place. Yet it would seem imperative that the potential nature and magnitude of these effects be understood and considered when applicable to field observations and measurements. It has been our experience that apparent manifestations of water aerosol absorption or emission seem to present themselves just as we are convincing ourselves that a previous observation of this effect must have some alternative explanation. In conclusion, we shall cite the most recent of these.

The Air Force has under development an infrared gas detector of the LOPAIR[8] type which is currently undergoing routine optical testing in Florida, near the Gulf of Mexico, in a transmitter-to-receiver configuration over a range of 2.3 miles, at an average elevation of the horizontal beam above the semi-arid surface of about 1 meter. This is an unusually long optical path for differential spectral measurements, and one would expect a plentiful supply of condensation nuclei in the area owing to the proximity to salt water and heavy aircraft activity. The system operates in the 8μ to 13μ window, using our natural-type optical filters.[32] While signal levels at the receiver are complicated by operational factors and a tendency toward optical mirage in looking over the line-of-sight, there is a tendency for plots of received signal versus relative humidity to follow the type of attenuation which would be predicted by our water aerosol theory even though visibility at 0.4μ to 0.7μ may not vary in the same manner. Figure A-12 shows a plot of many data points taken on 17 April 1969 from 0900 to 2315 hours. Despite the fact that air temperature and absolute humidity were very stable during this period, the trend of received signal, as approximated by the dashed line, is clearly in the downward direction with moderate increases in relative humidity, as shown in our original paper,[1] to lie in the region of potential exponential attenuation. In figure A-13 the curve showing the trend of data from the previous figure is compared with those for 2 other days, 18 April 1969 and 1 May 1969. The meteorological conditions for 18 April were very similar to those on 17 April, and the curve shows a similar but more exponentially-shaped trend. By comparison, the data for 1 May were taken under conditions of lower temperature with greater diurnal variation, and the absolute humidities were considerably lower than those for the first two curves. In this case, the received signal is much larger than in the previous trials, and shows no tendency toward attenuation at higher humidities which, in fact, went no higher than about 83% RH. We would suspect that comparatively few condensation nuclei were available here.

Steam emission experiments have been started at Edgewood Arsenal, utilizing the scanning radiometer described earlier in this paper. Preliminary spectra have shown strong emissions in the 8μ to 13μ region, presumably from water aerosol since the long-wavelength wing of the 6.5μ water band is also being monitored as an indication of vapor concentration. It has not been possible to explain the observed results using the distant wing theory. A paper describing these experiments and their potential impact will be forthcoming.

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APPENDIXES

APPENDIX A
FIGURES

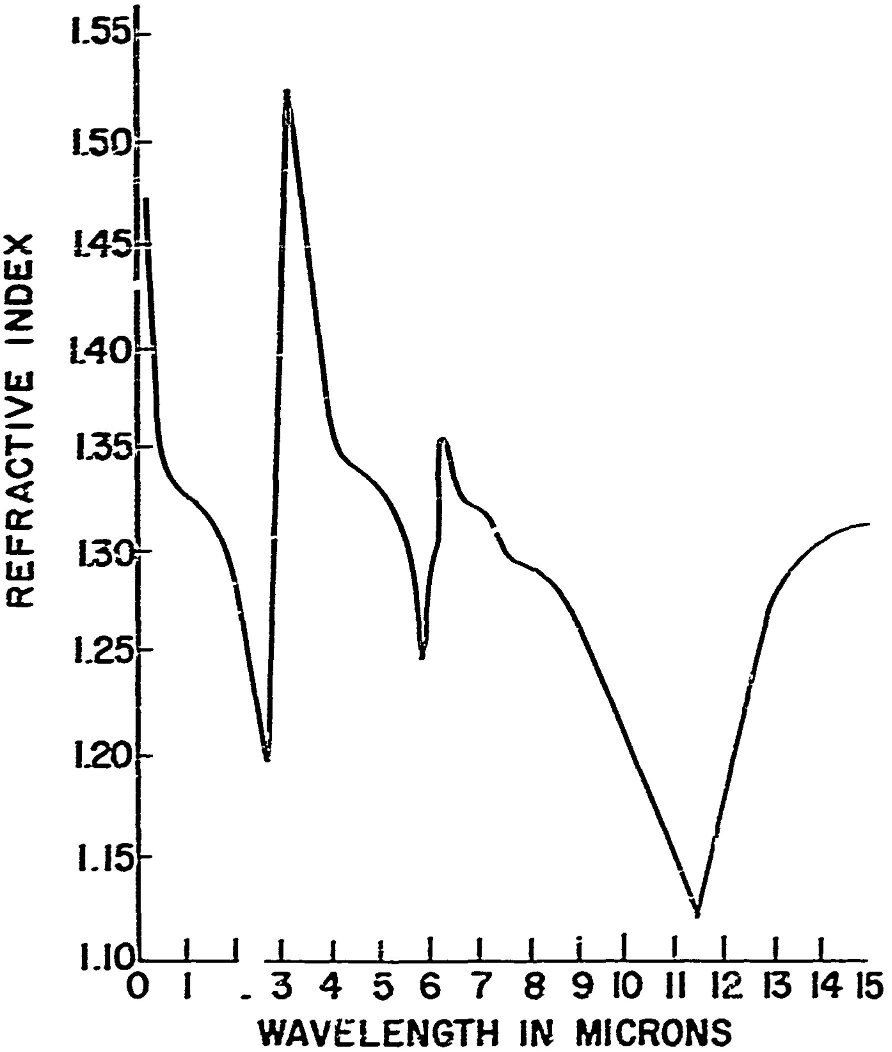

Figure A-1. Dispersion of Water in the Near Infrared

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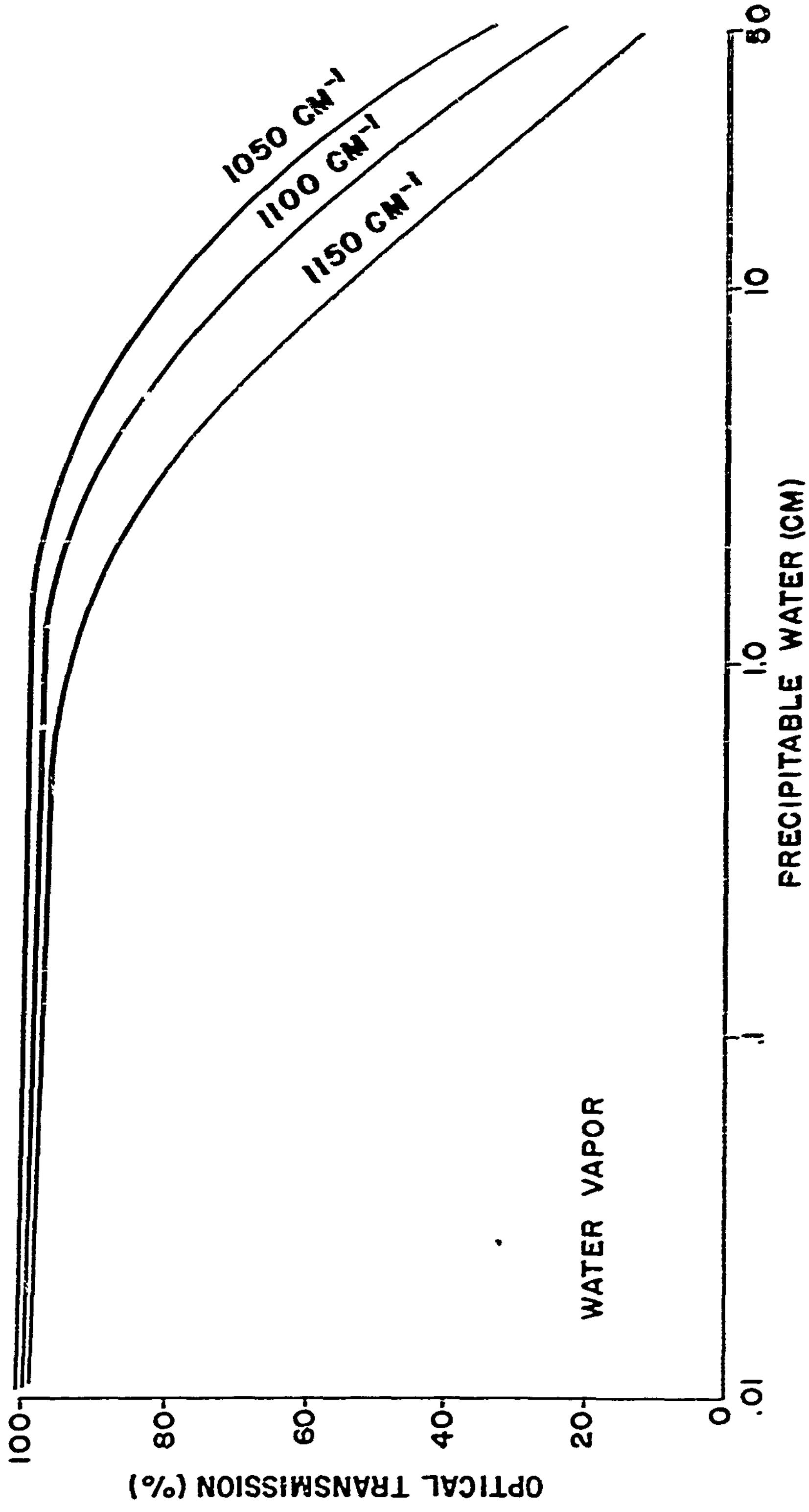


Figure A-2. Infrared Spectral Properties of Water Vapor

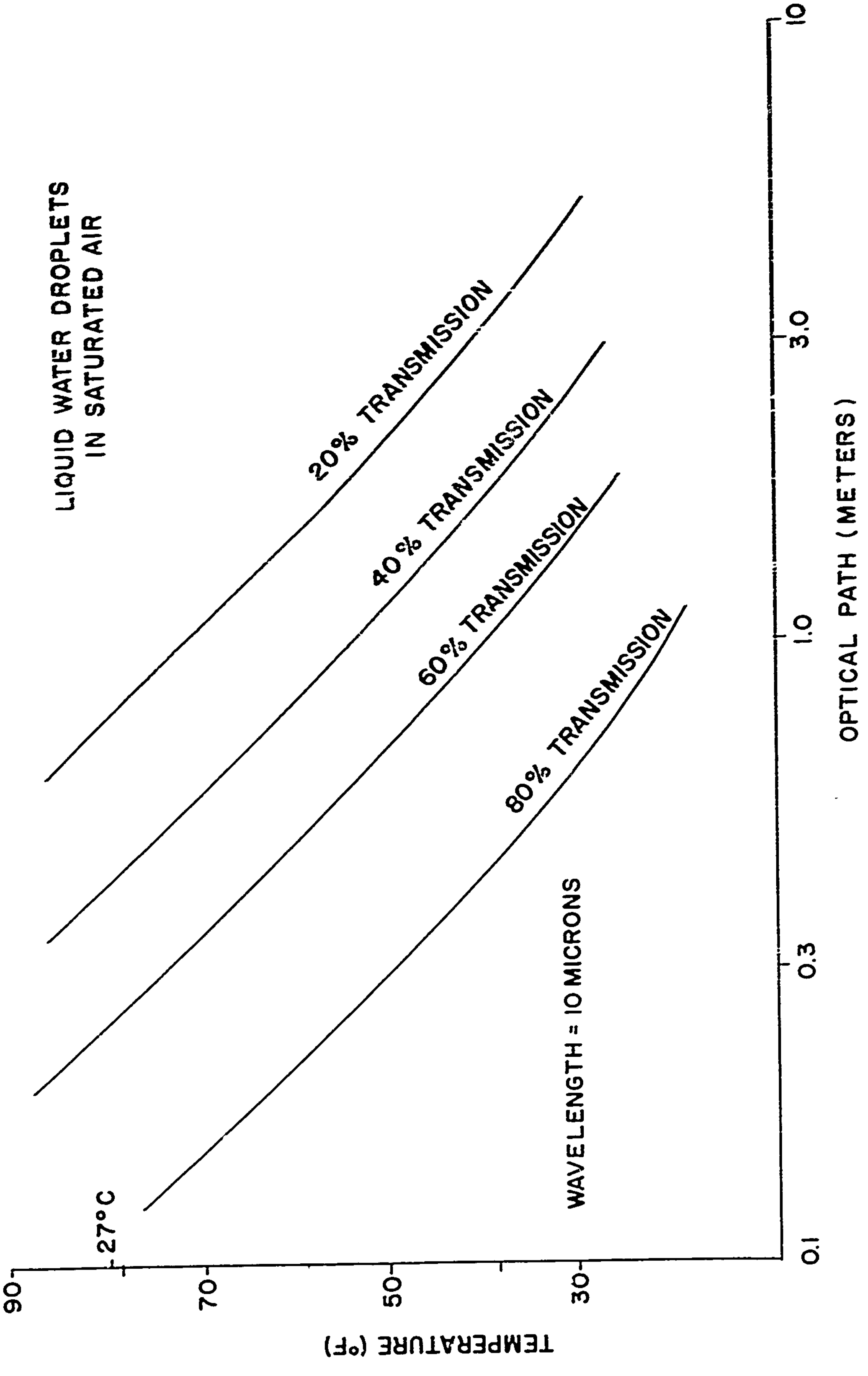


Figure A-3. Attenuation at 10μ by Nonscattering Fine Aerosol in Saturated Air

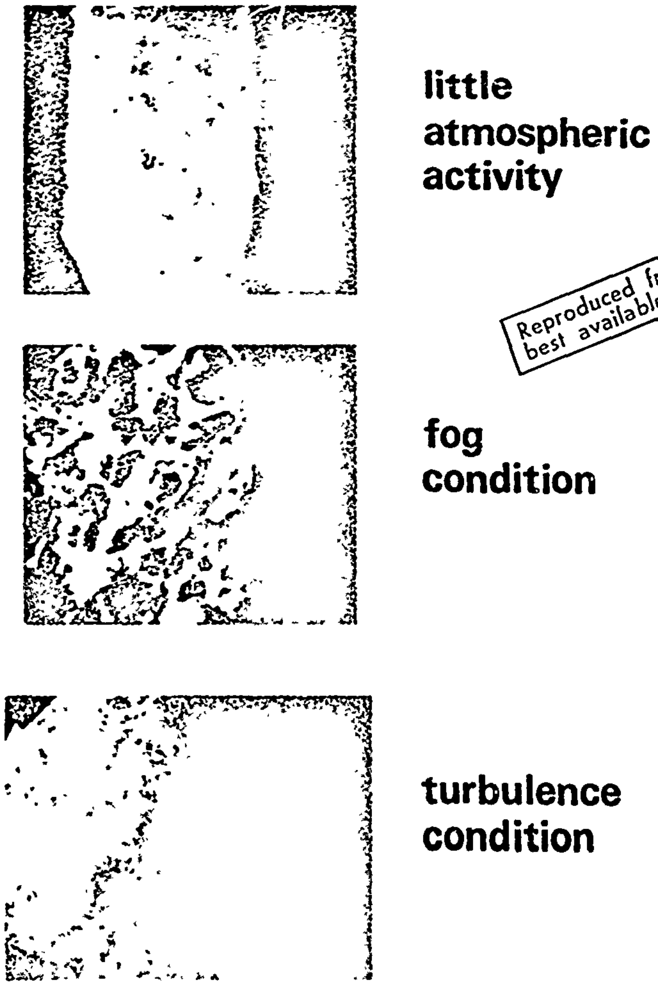
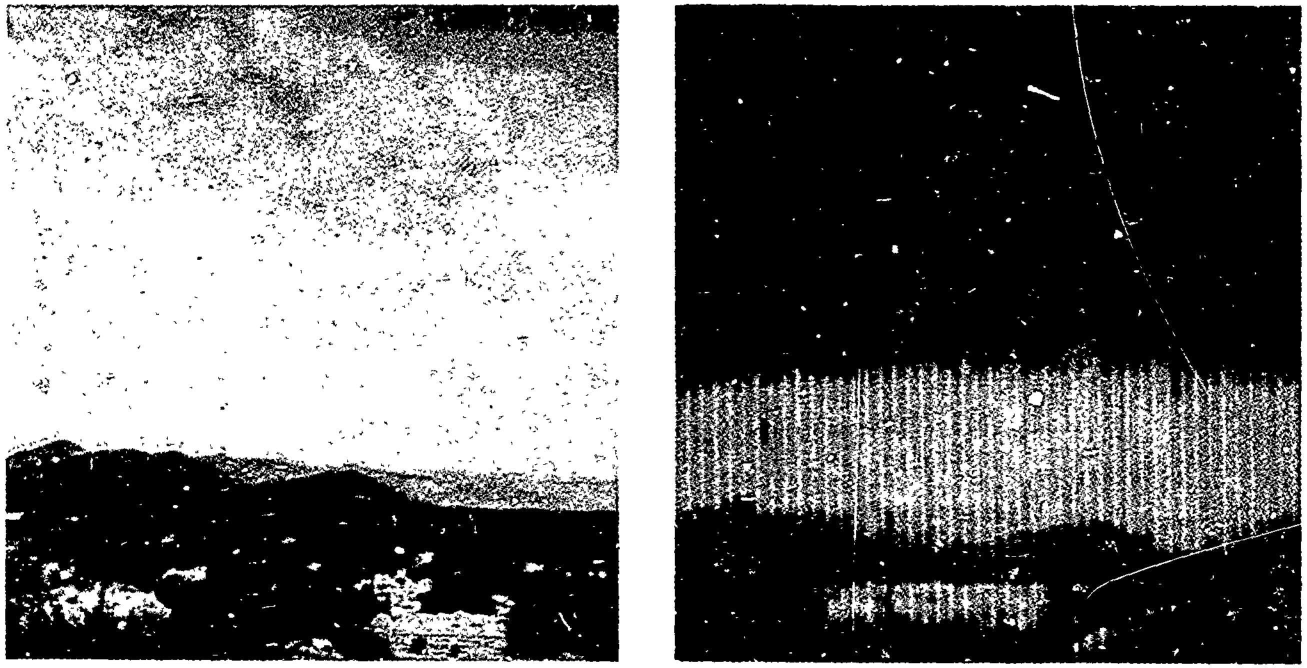


Figure A-4. Different Atmospheric Projection Phenomena



(a)

(b)

Figure A-5. (a) Photograph taken across the desert floor on a warm and somewhat humid day. (b) The air glows incandescent in the 8μ to 14μ band.

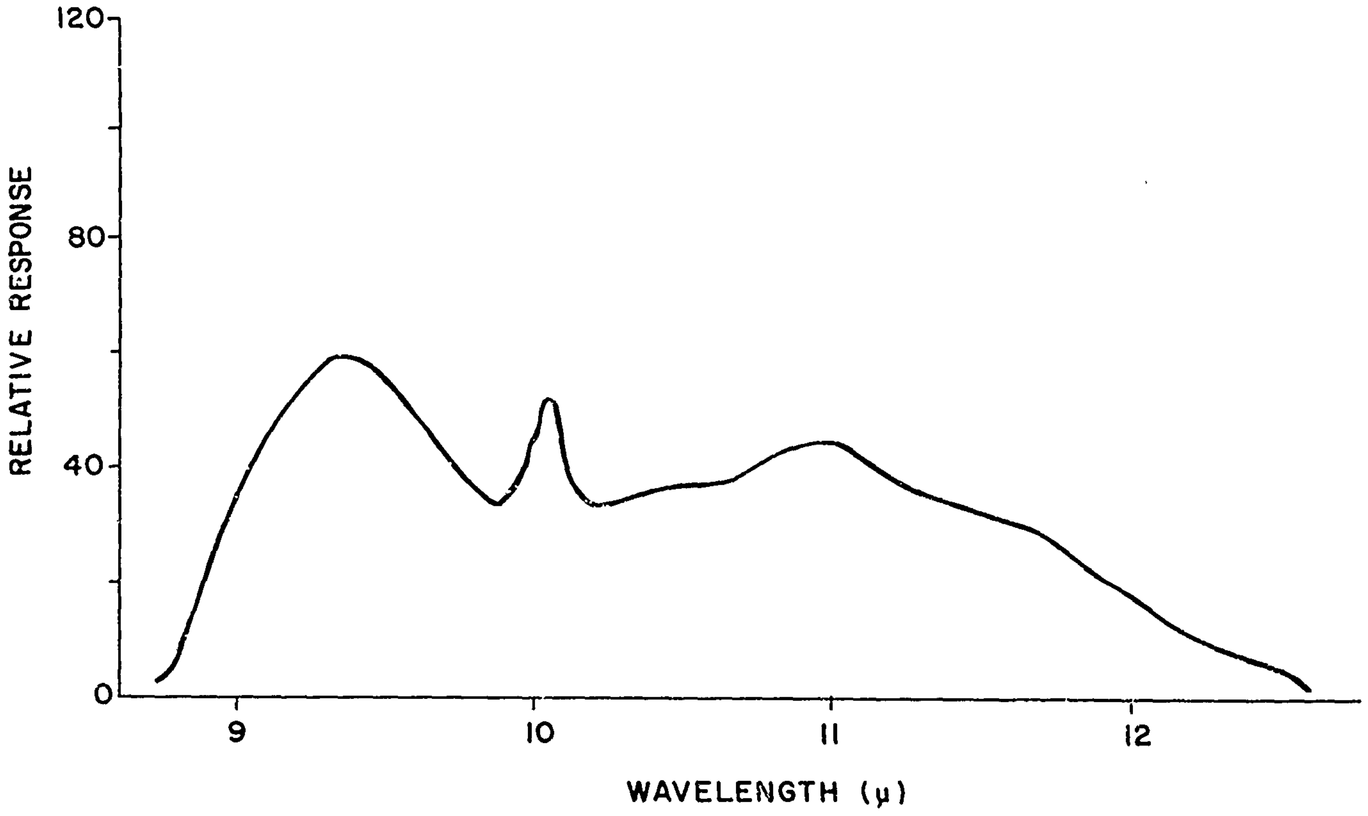


Figure A-6. Spectral Modulation Response of Instrument With Modified Filters

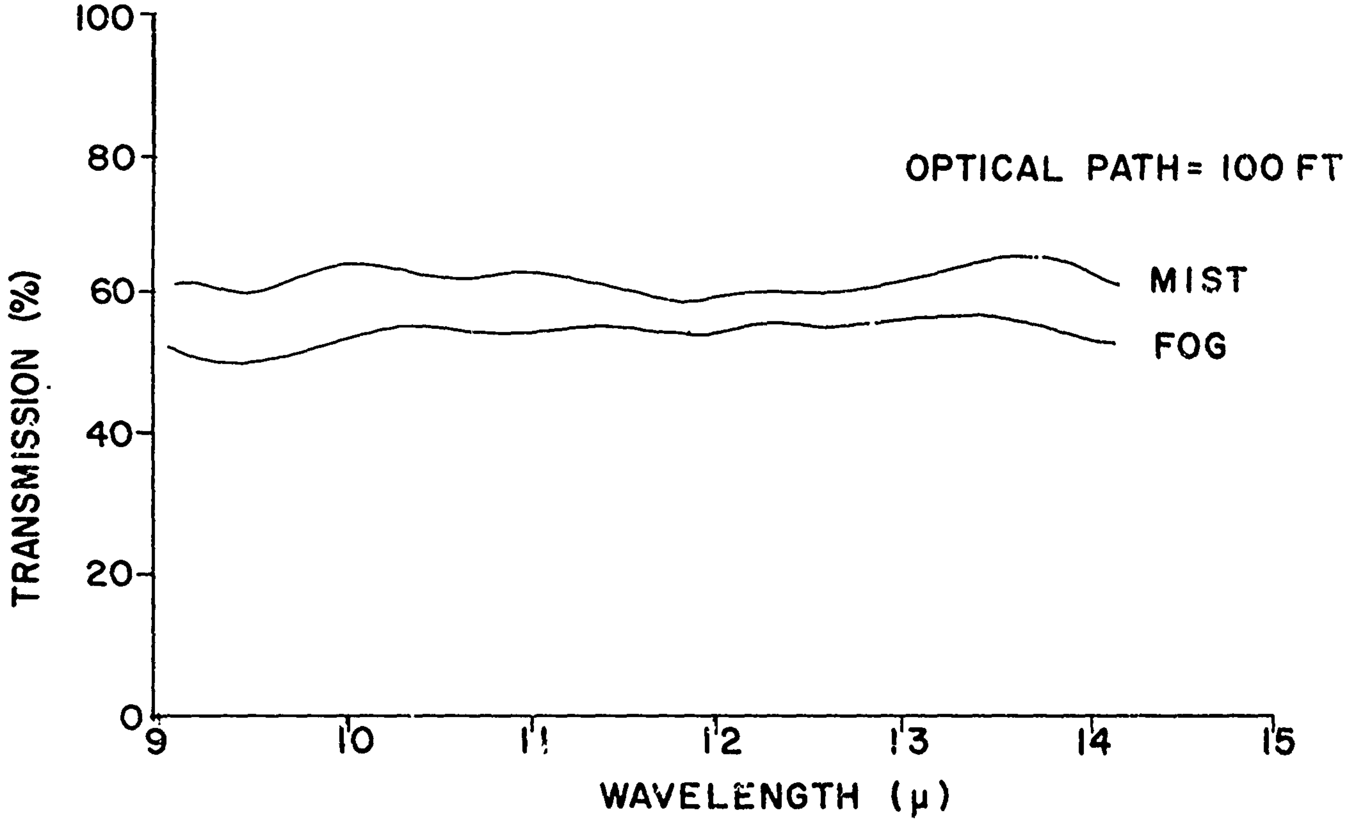


Figure A-7. Spectral Scans of Mist and Light Fog

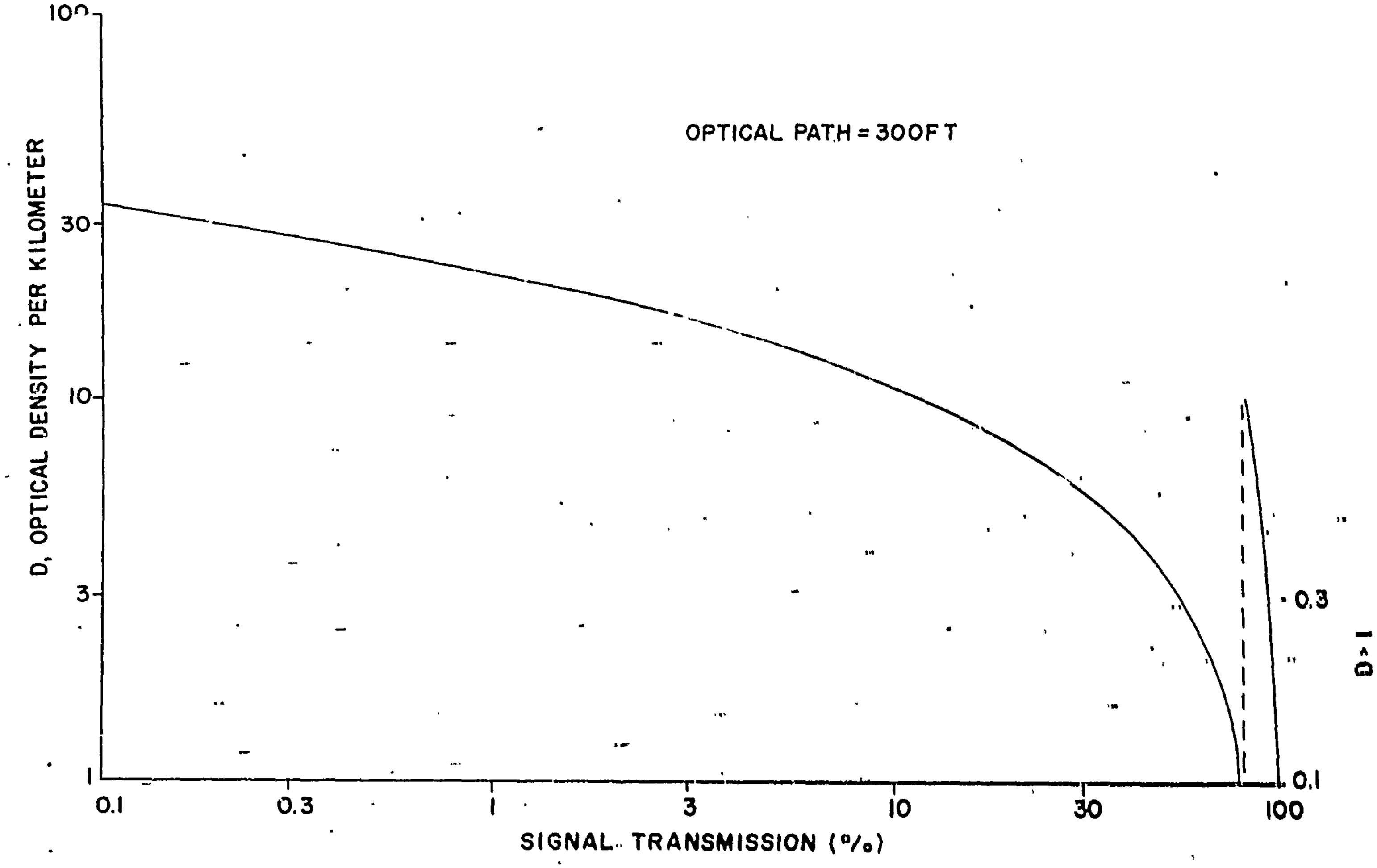


Figure A-8. Optical Density for Experimental Fog Measurements

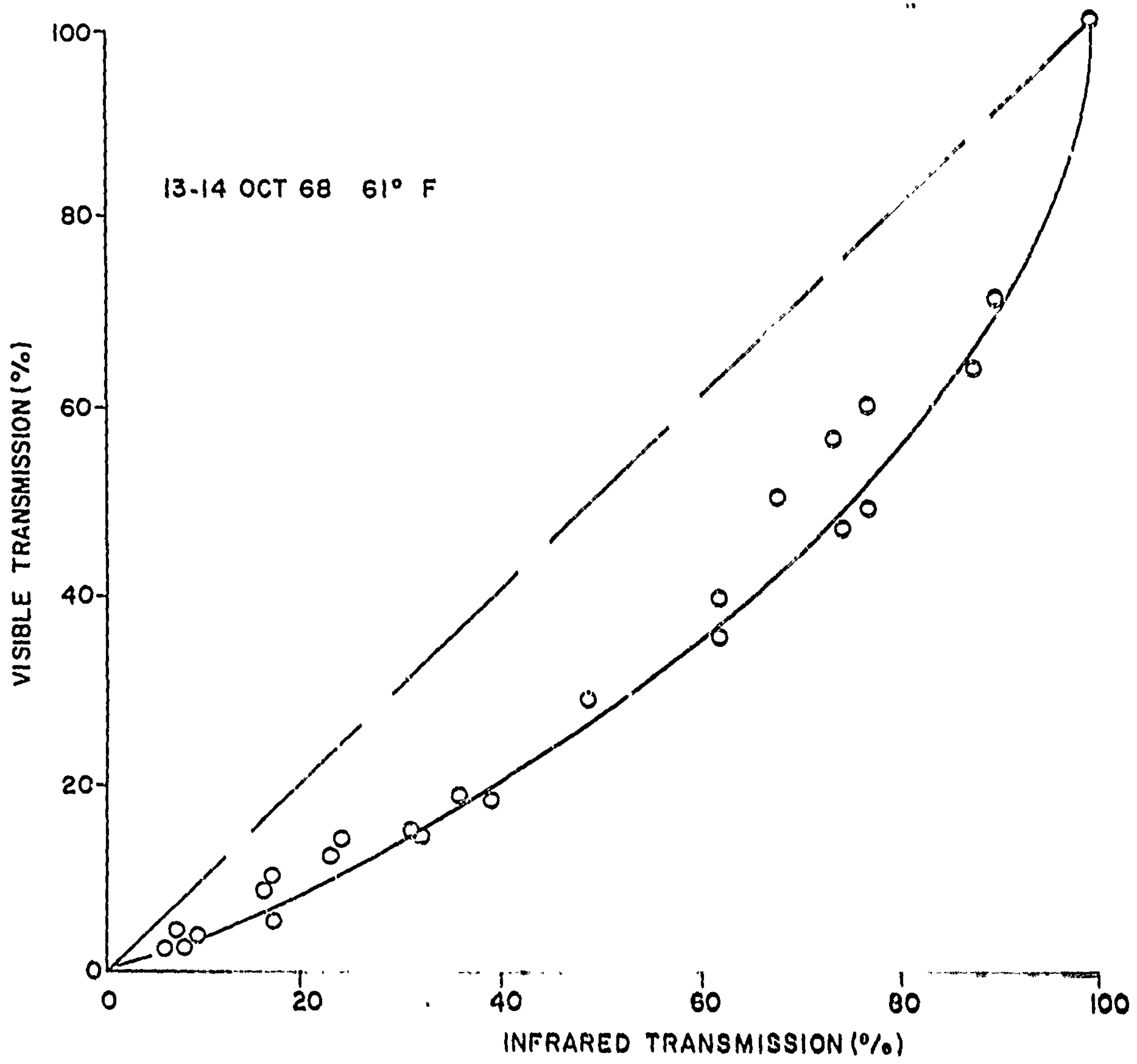


Figure A-9. Fog Trial (Optical Path = 300 ft)

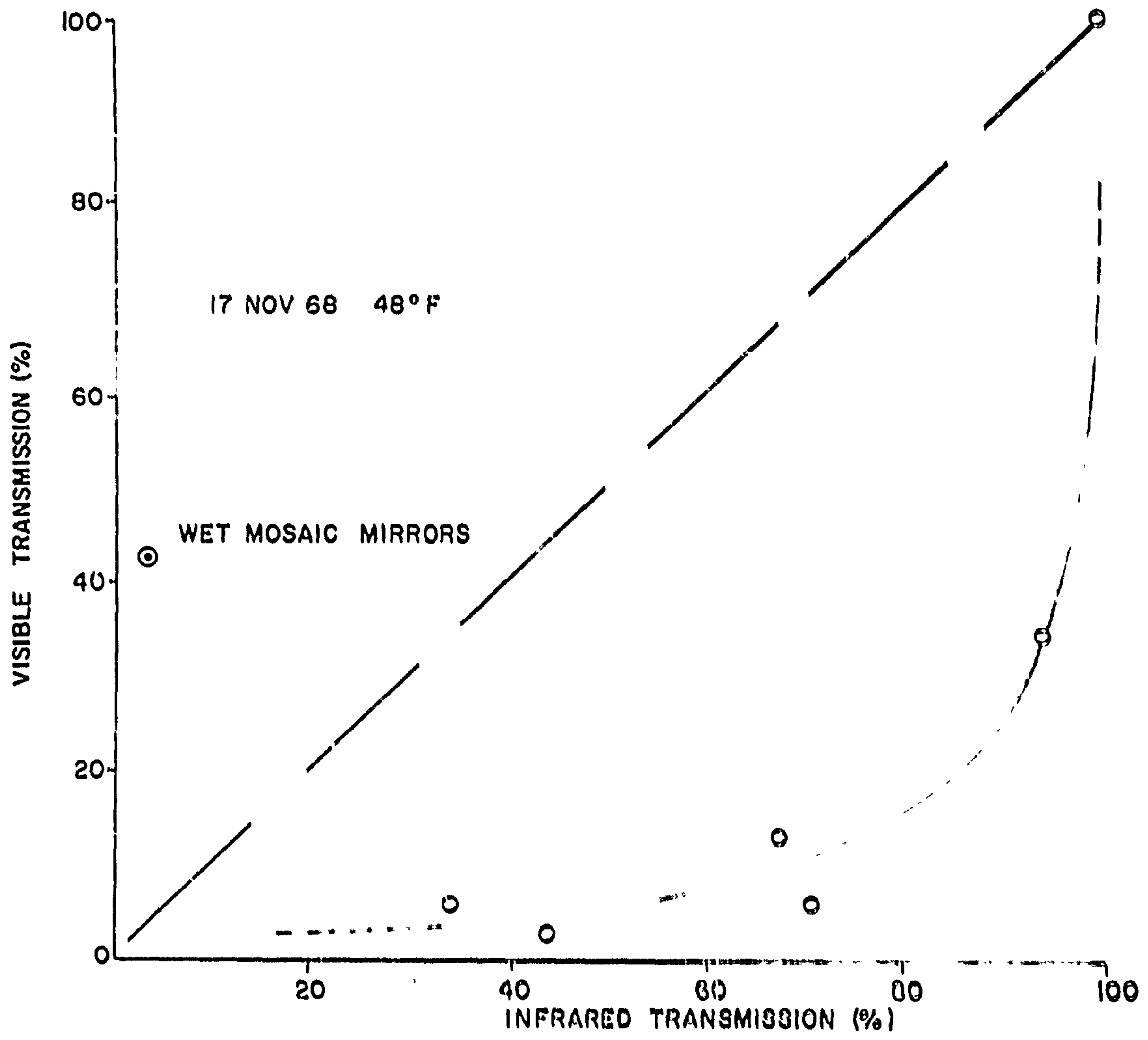


Figure A-10. Fog Trial (Optical Path = 300 ft)

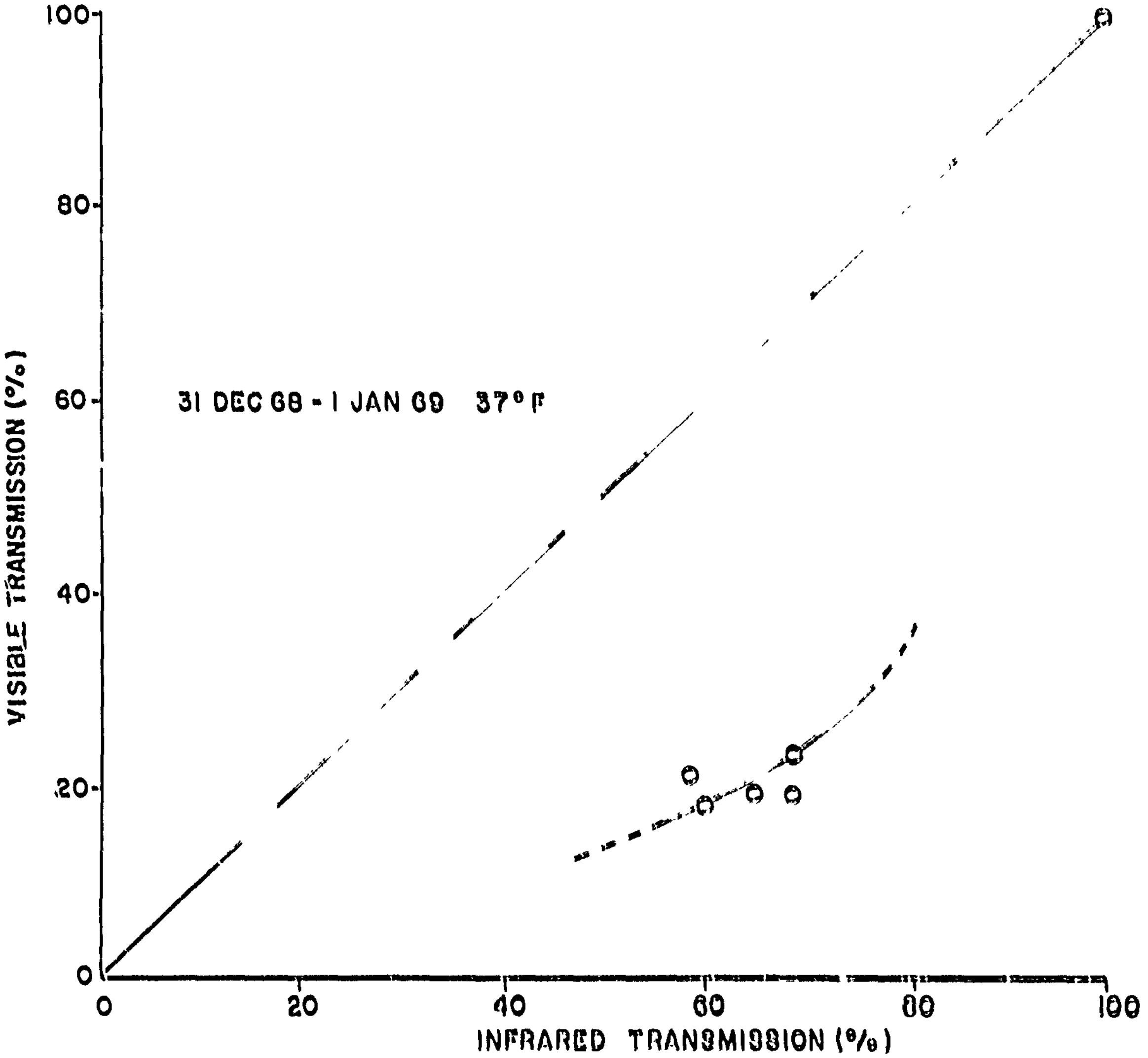


Figure A-11. Fog Trial (Optical Path = 300 ft)

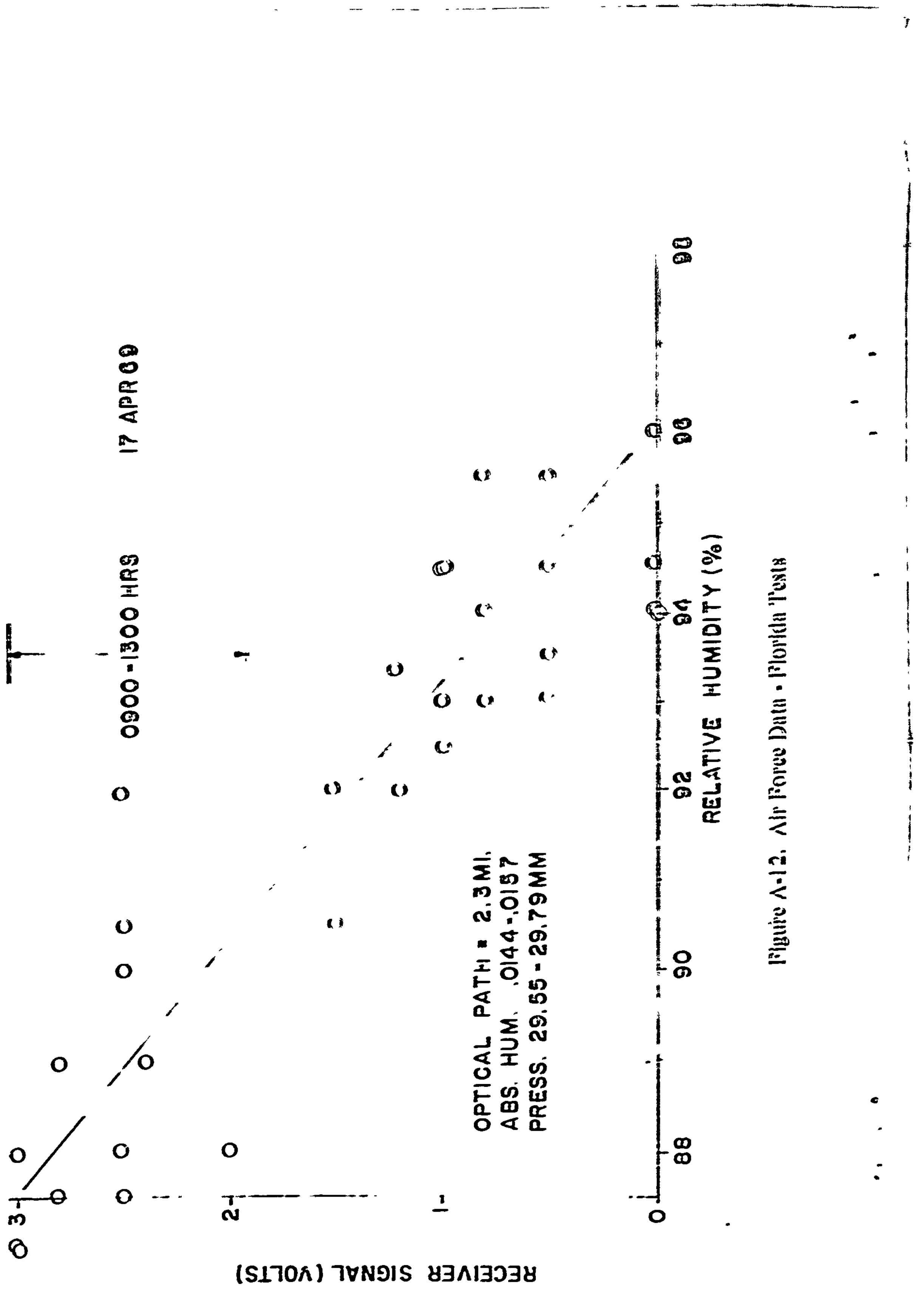


Figure A-12. Air Force Data - Florida Tests

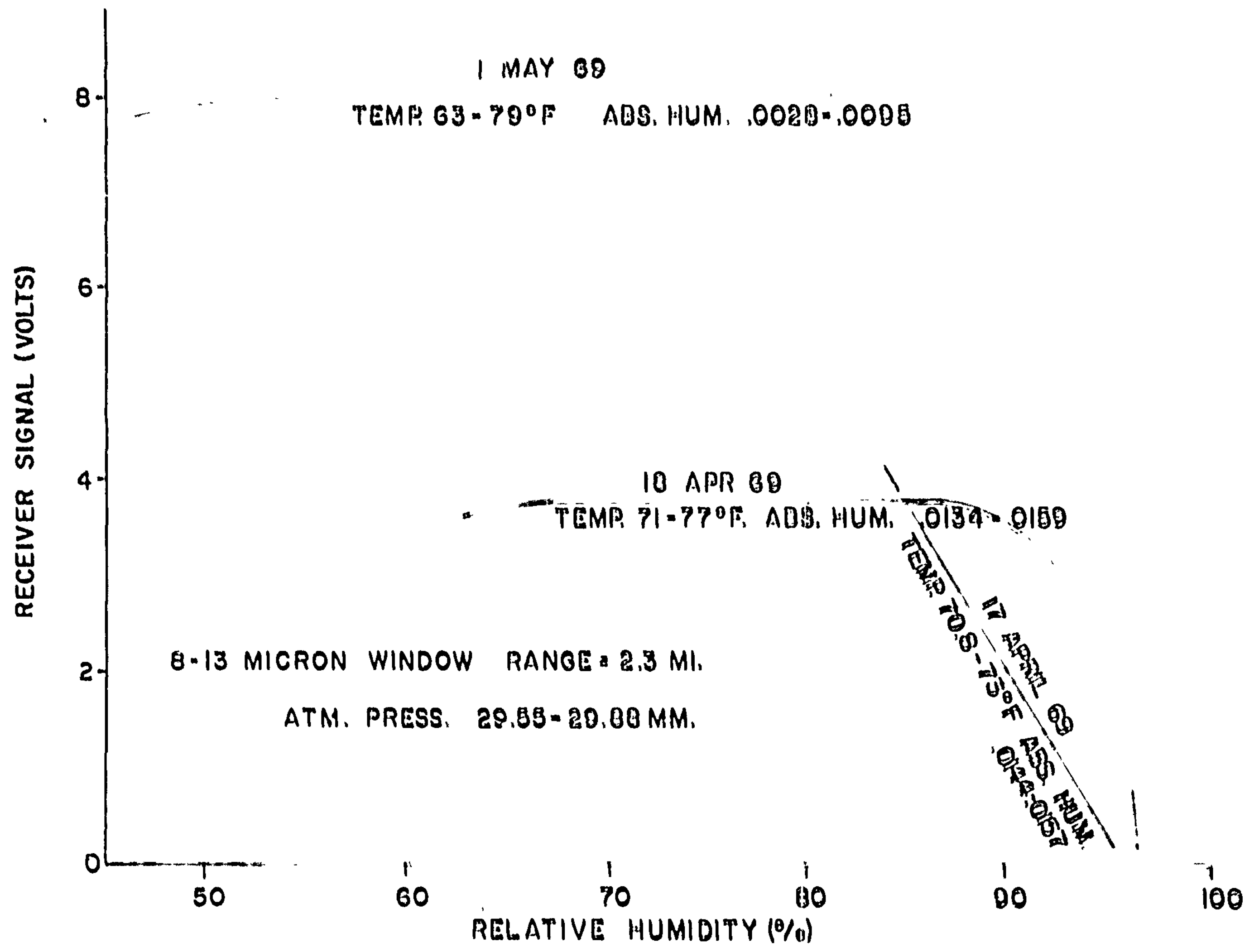


Figure A-13. Air Force Data Trends

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APPENDIX B

THE POSSIBILITY OF CLEAR-AIR TURBULENCE (CAT) DETECTION THROUGH HUMIDITY EFFECTS IN THE 8-13μ INFRARED WINDOW

by

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In a previous publication,[1] the author mentioned the possibility that humidity effects in the 8-13μ infrared window region might permit detection of clear-air turbulence (CAT) ahead of an aircraft. This detection would be based upon the indication that water aerosols in equilibrium with water vapor at higher relative humidities can cause very significant optical attenuations in this spectral region, since water, in going from the vapor to the liquid phase, undergoes an increase in optical density of three to four orders of magnitude. This approach was fully developed in an earlier paper which was concerned with possible humidity effects upon terrestrial scintillation intensity.[2] In that paper it was stated that, while condensation nuclei were necessary for the formation of these aerosols, the droplet radii could be very small ($10^{-2}\mu$) without reducing the magnitude of attenuation. A recent review article on clear-air turbulence[3] states: "The most substantial indicators of air motion aloft, however, are the water-substance particles of clouds, and also the particulate matter present in "clear" air. Apart from the more obvious forms of high-level water-substance cloud, there is increasing evidence that sub-visible stages of such clouds exist and that haze or dust concentrations are very variable." Thus, we see that, in apparently "clear" air, both the condensation nuclei and resulting sub-micron droplets necessary to attenuate in the 8-13μ infrared window may be present.

While it is obvious that high humidities may be readily associated with low-altitude convective columns or rising air currents over mountain ranges, the occurence of high-humidity gradients at typical CAT altitudes is open to question, especially since optical attenuation would depend first upon relative humidity for formation of water droplets, but, second, upon specific or absolute humidity. At altitudes of 20,000 feet, temperatures are low enough to prevent the incidence of absolute humidities greater than a small fraction of those at the earth's surface. For that matter, there is no guarantee that ice crystals might not be more likely to be encountered than water droplets. In addition, there is the question as to whether humidity gradients at these altitudes would show correlation with CAT incidence. It was apparent from the selection of papers presented at the recent National Air Meeting on Clear Air Turbulence[4] that no approach to the problem of CAT detection can be overlooked. Many of these approaches seem to rest on much less substantial foundations than the techniques proposed here.

Specifically, it would be necessary to optically scan the area ahead of an aircraft with an infrared system similar to one already proposed to monitor temperature gradients[5] using the CO_2 absorption band near 15μ. In this case, however, a narrow or fixed wavelength band would be used between 10μ and 12μ. If the scan were ahead and up (against sky background), humidity gradients should appear as emitters on a cold background. If the scan were ahead and down (against earth low-altitude background), these gradients might appear as absorbers on a warm background. In either case, they should provide a dynamic picture of turbulence activity when necessary conditions are met. The selection of an instrumented or visual readout would have to be made on the basis of performance data for such a system.

At least one major instrument company has expressed an interest in proposing to interested agencies a scanning system based upon these concepts. The author feels that such a proposal would be a worthwhile undertaking, and welcomes comments concerning this technical approach to the CAT problem.

HUGH R. CARLON
April 21, 1966

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APPENDIX C

INFRARED APPLICATIONS BRANCH
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EFFECTS OF HIGH HUMIDITY UPON ATMOSPHERIC TRANSMISSION AND INTERPRETATION OF RADIOMETRIC TEMPERATURE MEASUREMENTS IN THE 8-13 MICRON IR WINDOW

In previous publications[1,2] the author has discussed in detail the potential effects of high-humidity air masses (*) upon the interpretation of radiometric measurements of the earth's temperature from orbiting satellites. It was suggested that at relative humidities of about 70% or more, water aerosols which may exist in equilibrium with water vapor can begin to absorb strongly far-infrared radiation, most particularly in the 10-12 micron region of the 8-13 micron atmospheric window (Window VIII). Although these air masses are essentially "invisible," they may seriously lower satellite temperature readings when they occur at higher altitudes under apparently clear conditions. The magnitude of this effect depends, of course, upon total water content (precipitable water) in the humid air mass.

Saiedy and Hilleary[3] have presented data which can be interpreted, using this water aerosol theory, in an alternative way to that which they present. In a series of balloon measurements of earth temperature at an 11.1 micron wavelength, taken from an altitude of 30 km, they experienced differences of as much as 8°C between calculated surface temperatures and radiometrically-measured temperatures under humid atmospheric conditions which they describe as "extreme". (It is the author's contention that such conditions are not as extreme as normally thought, and that temperature data of this type are often misinterpreted for this reason). In Figure 5 of their paper, Saiedy and Hilleary show the progressive growth of this temperature difference as the day proceeds. In early morning, the difference is only 2°C. By 11:00 a.m., the 8°C variance is observed. Then, within an hour, a large cloud mass obscures observation of the earth's surface, and a thunderstorm ensues. This behavior is precisely that which would be expected according to the water aerosol theory, and could be explained as follows. As the humid air mass moved into the test area, accompanying increases in relative humidity at altitudes below the observation balloon began to attenuate the 11.1 micron measurement wavelength, although the optical path was apparently clear. Nascent clouds, at temperatures well below those at the earth's surface depending upon their altitudes, began to drive the radiometric temperature in the predictable, downward direction. If measurements had been continued between 11:00 a.m. and 12:00 Noon, when the thunderstorm began, it is likely that the temperature measurement difference would have become still greater. In fact, this difference would grow exponentially according to theory. During the measurement period cited above, a precipitable water content of 5.4 cm was noted. By comparison, another reported test was run under conditions such that the precipitable water content was only 0.5 cm. It is worth noting in the latter test that virtually perfect agreement was obtained between radiometric and actual earth surface temperatures, although some instrument problems were encountered.

(*) The term "air mass" as used in this correspondence is intended to imply a patch or nascent cloud of high humidity content, rather than a widespread body of air as is perhaps a more commonly-understood interpretation.

A recent paper[4] gives an excellent description of the formation of these water aerosol masses, as previously theorized, and their effects upon radiometric measurements of the temperature at the surface of the sea. Clark, the author, states "when a layered structure exists, extrapolation (of radiometric data) is not practicable, particularly when the layer is composed of saturated air close to the surface of the sea. In such layers, may be found nascent clouds and those water vapor concentrations which eventually develop into ground fog. Such concentrations may cover several hundred square miles and, since they are invisible, may be present without the knowledge of the airborne observer, thus leading to errors of from a few tenths to several degrees. Of course, once the low hanging clouds or ground fog has developed, an opaque layer is presented to the radiometer which prevents further viewing of the sea."

The humidity effects responsible for apparent discrepancies while measuring with a radiometer through vertical or slant paths are also pronounced in the operation over horizontal paths of "line-of-sight" optical systems. Although the latter are usually self-limiting with respect to path length owing to terrain features, it has been shown[1] that high-humidity conditions can cause serious attenuation of infrared signals in the 8-13 micron window, in regions which are supposed to be highly transparent, and that these conditions are also believed to contribute to the intensity of terrestrial scintillations at relative humidities above 70%. These effects may well account for the difficulty in establishing reference levels encountered by workers measuring atmospheric spectra as reported, for example, by Taylor and Yates.[5] A recent paper[6] notes that new transmission data obtained in Window VIII were not in agreement with earlier measurements cited in Reference 5, under such circumstances that "the only significant difference in the meteorological conditions under which the spectra were obtained is that of *relative humidity*" (the italics are those of the undersigned). Reference 1 is then cited as a possible explanation.

Studies of the effects upon atmospheric transparency of varying high humidity levels, most particularly in the 8-13 micron window, are continuing at Edgewood Arsenal. The shape of the transmission characteristic of this window with changing humidity should be studied in detail since theory predicts that water aerosols more sharply attenuate the 10-12 micron region than other wavelengths. This information would be directly extrapolable to vertical and slant-path situations as well. Additional findings will be reported in future papers.

HUGH R. CARLON

AD-753 347

TUNABLE PARAMETRIC OSCILLATORS BETWEEN 1.0 MICROMETERS - 5.0 MICROMETERS

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October 1972