gives the x-ray diffraction pattern of rhombic sulfur. On the other hand, the product quenched from above our liquidus (agreeing in general with the liquidus reported by several workers) and examined immediately is a viscous liquid. Moreover, according to Tammann (*12*), the rhombic-monoclinic-liquid triple point occurs at 1294 bars and 151°C, and the monoclinic-liquid equilibrium boundary has a positive slope.

In addition to the evidence on stability fields for 12 crystalline high-pressure forms of sulfur, we have identified four different high-pressure sulfur liquids below 30 kb (*13*); data for our melting curve have been compared with those of other investigators (*14*).

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Galactic Water Vapor Emission: Further Observations of Variability

Abstract. *Recent observations of the 1.35-centimeter line emission of water vapor from galactic sources show short-term variability in the spectra of several sources. Two additional sources, Cygnus 1 and NGC 6334N, have been observed, and the spectra of W49 and VY Canis Majoris were measured over a wider range of radial velocity.*

After the discovery of the 1.35-cm line emission of water vapor from four galactic sources (*1*), observations made with higher telescope resolution (*2*) showed intense radiation from eight galactic source regions of small diameter. Pronounced variations in the spectrum of the source region associated with W49 were observed over a few weeks' time (*2*). The observation of such short-term variability is critical to the understanding of the regions of water vapor emission and implies a very small diameter for the emitting region. We have made new observations between 15 March and 9 April 1969, using the 85-foot (26-m) reflector at the Naval Research Laboratory's Maryland Point Observatory and spectral-line radiometer system described in our previous paper (*2*). Spectra were measured with a spectral resolution of 4 khz except for the very wide spectrum of W49 (Fig. 1), which was measured with a resolution of 100 khz. The level of fluctuation in random noise had a root-mean-square value of 10°K for the 4-khz spectra and 3°K for the 100-khz spectrum. Additional fluctuations due to the gain calibration with a noise source of 90°K are present in strong spectral features. The observed antenna temperatures have been corrected for atmospheric absorption ranging from 10 to 30 percent and are accurate to about 10 percent except for the noise-limited weak features. No correction has been applied for changes of antenna gain with zenith angle; this would affect significantly only those sources at large negative declinations. This correction is not accurately known, but it may be as large as 15 to 20 percent at a declination (δ) of −30°. Unless otherwise specified, the observations were made with linear polarization at a position angle of 0°.

The observations show further time variations in the spectrum of W49, variations in the spectra of the source region associated with the Orion Nebula, the thermal radio source G133.7 + 1.2 (W3), and W75S, and a probable change in the spectrum of G0.7 − 0.0 (Sagittarius B2). Water vapor emission was observed from the OH emission source, Cygnus 1, discovered by Ellder *et al.* (*3*). We have also observed H_2O emission from NGC 6334N and from the additional feature at + 37 km/sec in the spectrum of VY Canis Majoris, both of which were observed by Meeks *et al.* (*4*).

All radial velocities reported by Knowles *et al.* (*2*) were referred to a nominal laboratory frequency of 22235.22 Mhz as given in the National Bureau of Standards tables (*5*). The results given here are referred to a probably more accurate rest frequency of 22235.08 Mhz, which is the weighted mean of the frequencies of the hyperfine components of the transitions from the 6_{16} to 5_{23} levels as measured by Bluyssen *et al.* (*6*). The radial velocity scales presented here are therefore shifted by 1.9 km/sec toward more positive velocity than those in the previous report (*2*), and the radial velocities of the observed components appear shifted by 1.9 km/sec toward more negative velocity than those reported earlier (*2*).

Typical spectra of the source region associated with W49, measured at intervals of about a month from 19 January to 6 April 1969, are shown in Fig. 2. The two intense lines at + 7 and + 14 km/sec, which disappeared between 25 January and 8 February 1969, have not reappeared. However, during late March and early April an intense line developed at + 9 km/sec, the same velocity as the third feature which disappeared in early February. In addition, the features at radial velocities of − 6 and + 25 km/sec have both decreased in intensity while others have increased. Two pairs of lines, the (+ 7 km/sec, + 14 km/sec) pair and the (− 6 km/sec, + 25 km/sec) pair, change intensity together and are roughly symmetrical about the strong feature at + 10 km/sec. This suggests some sort of association involving the components of the pairs and the possibility of ordered motions of the emitting gas clouds. In our previous paper (*2*) we mentioned that the spectrum of the W49 source extended to a radial velocity of − 150 km/sec. This finding was confirmed by Meeks *et al.* (*4*). A wide-range spectrum of the W49 source measured with 100-khz resolution in early April is shown in Fig. 1. We did not observe the intense feature at + 140

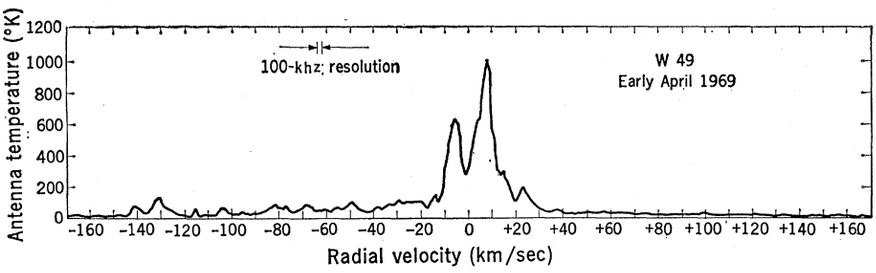

Fig. 1. Spectrum of the H_2O source associated with W49 measured with 100-khz resolution; significant water vapor emission spread over a wide range of radial velocity to — 140 km/sec is shown.

km/sec reported by Meeks *et al.* (*4*); this result is apparently in agreement with their conclusion that the feature at + 140 km/sec disappeared between 16 February and 9 March. The features at + 140 km/sec and — 130 km/sec may provide additional evidence for symmetry. The variable feature at + 9 km/sec and the feature at + 10 km/sec are separated in frequency by 80 ± 10 khz in the January spectra and by 70 ± 10 khz in the March-April spectra. Within experimental error this separation is in agreement with the difference in frequency of 76.2 khz (*6*) associated with the hyperfine transitions from the 7 to 6 and 5 to 4 levels; thus the observed frequency difference may be due to hyperfine transitions rather than to two emission regions at different radial velocities.

The most intense feature in the W75 source region, at a radial velocity of + 3 km/sec, decreased in antenna temperature from more than 300°K to about 20°K between 16 February and 8 April, as shown in Fig. 3. The weaker line at 0 km/sec maintained about the same intensity. This provides evidence in a second source region for a drastic change of intensity in a short time; the size implied for the emitting region in this case is less than 0.03 parsec. As in the case of the W49 source, polarization measurements rule out the possibility that the observed variability is simply a change of the state of polarization of the received wave.

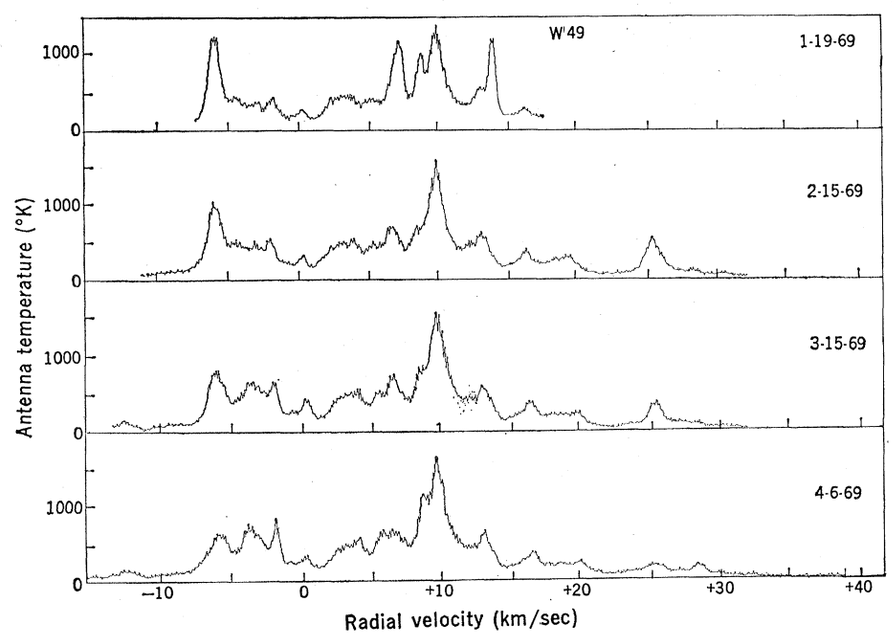

Fig. 2. Central portion of the water vapor spectrum of the source associated with W49 measured with a resolution of 4 khz at intervals of approximately 1 month; time variations are shown. The spectrum of 19 January 1969 was measured in rain, which accounts for the reduced amplitude. The velocity scales mentioned here are based on a more accurate rest frequency than used previously (*2*).

The feature in the Orion spectrum at + 0.5 km/sec is unique among presently measured H_2O sources because of its high polarization (30 percent linear). During our January-February observing period the feature at + 0.5 km/sec appeared clearly distinct from a weaker line at — 1 km/sec; the characteristic shape of this spectrum persisted through our last data observed on 14 February. Spectra taken between 22 March and 9 April at a position angle of 0° show the feature at + 0.5 km/sec to be broadened and shifted toward negative radial velocity, merging with the feature at —1 km/sec, as illustrated in Fig. 3. This suggests as an alternative that the feature may be regarded as two displaced components, with the one at more negative radial velocity increasing in intensity while the one at more positive radial velocity is decreasing. The measured frequency displacement of the feature which increased in March from that in January is 40 ± 10 khz, in good agreement with the splitting between the hyperfine transitions from the 6 to 5 and 5 to 4 levels; this again suggests that a change in the hyperfine components rather than Doppler-shifted components may be a satisfactory alternative interpretation for the observed frequency difference. A spectrum taken with 90° polarization on 27 March shows the peak still at the same frequency as in January to within 5 khz, an indication of a different preferential plane of polarization for the two components.

Because of an error, the antenna temperature scale for the Orion spectrum in Fig. 2 of our previous paper (*2*) is too large by a factor of 2 [erratum (*2*)].

The water source associated with the W3 continuum source G133.7 + 1.2 was measured several times during March and April and was found to decrease gradually in intensity. The antenna temperatures of the more intense features around — 41 km/sec measured in early April were about 60 percent of those measured in January-February (*2*), whereas the intensity of the weak feature at — 36 km/sec remained nearly the same. No significant changes were found in the spectrum of the water source associated with the W3 hydroxyl source.

The spectrum of the weak line emission from Sagittarius B2 was measured again in March. The March spectra show a single feature with an antenna

temperature of 60°K and a radial velocity of + 69 km/sec, in agreement with the February spectra of Meeks *et al.* (*4*). A feature observed in January at + 36 km/sec (*2*) was not observed in the March spectra. Since we measured the spectrum only once during the month of January, we do not have confirmation of this feature and can only list this source as probably variable.

A new H_2O source region was found in the direction of the OH emission source Cygnus 1 recently discovered by Ellder *et al.* (*3*) in their search for OH emission regions not associated with known continuum sources. The main feature of the H_2O line emission spectrum has an antenna temperature of about 30°K and is centered at a radial velocity of + 16 km/sec; a weaker feature is observed at + 10 km/sec. Ellder *et al.* (*3*) found OH emission features at a radial velocity of between + 13 km/sec and + 17 km/sec. The observed position of the H_2O source is α_{1950} (right ascension) = $20^h08^m00^s \pm 4^s$, $\delta_{1950} = 31°22.4' \pm 1'$. As pointed out by Ellder *et al.* (*3*), there is no apparent H II region at this position, but there are several highly reddened stars in this heavily obscured region. No H_2O line emission with antenna temperature greater than 10°K was detected from the three other OH emission sources discovered by Ellder *et al.*

We have also observed the previously reported (*4*) H_2O emission from the position (*7*) of the northern OH emission source associated with NGC 6334. The measured position of the H_2O source is $\alpha_{1950} = 17^h17^m31^s \pm 8^s$, $\delta_{1950} = -35°44' \pm 2'$. The observed H_2O line emission has an antenna temperature of about 25°K and is centered on a radial velocity of − 6 km/sec, where a broad OH absorption is observed, rather than on a velocity associated with OH emission features (*8*). The H_2O velocity is close to the radial velocity of − 5.1 ± 0.7 km/sec determined for the H II region from observations of recombination lines (*9*).

No significant changes were found in the spectra of the W51 and VY Canis Majoris sources. Measurement of the spectra over a wider frequency range disclosed an additional relatively weak feature with an antenna temperature of 100°K in the spectrum of VY Canis Majoris at a radial velocity of + 32 km/sec. This feature, which has also been observed by Meeks *et al.* (*4*), is toward the negative-velocity side of the broad-band OH emission feature (*10*) which extends from + 32 to + 60 km/sec.

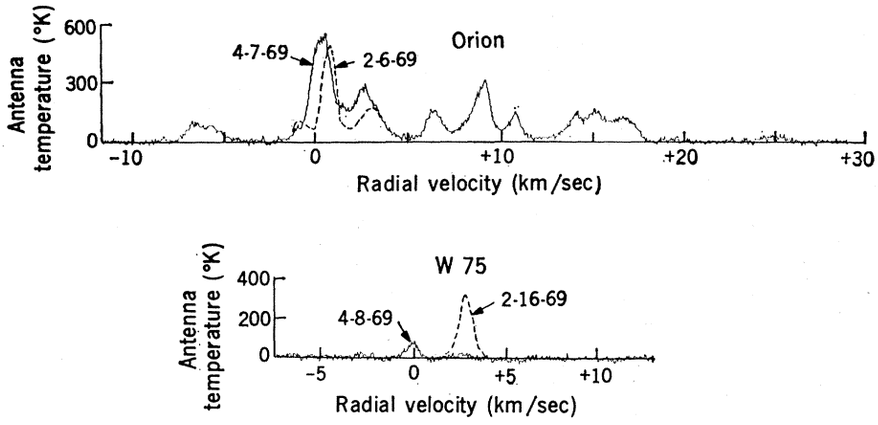

Fig. 3. Composite spectra of the H_2O sources in Orion and W75 measured with 4-khz resolution; time variability is shown.

The observation of intensity variations with a period of 1 month or less in several water vapor sources shows that this is not an isolated phenomenon in these regions. These variations along with the implied high brightness temperatures permit estimates of some properties of the H_2O emission regions and the emission process.

If we assume that the observed radiation is the result of maser amplification (*2*), intensity changes may be related to changes in maser gain. The relation between the input intensity (I_i) and output intensity (I_0) of the radiation of a maser amplifier can be written as

$$I_o = I_i e^{(\alpha L)}$$

where (αL) is the maser gain constant. We may assume that a cloud of water vapor is illuminated by radiation with a brightness temperature of a few degrees Kelvin, from the 3°K isotropic blackbody radiation and the continuum emission from the nearby H II region. Based on the estimated upper limit to the source size derived from short-term variability, the strong features in the W49 spectrum with a brightness temperature of more than 10^9 °K (*2*) would require a gain constant (αL) of at least 19.

Several important properties of the maser amplification depend on whether the maser is saturated, that is, whether the stimulated microwave transition rate is as fast as the net pumping rate which maintains the population inversion. Using Eq. 4 of Litvak *et al.* (*11*) with the rate of spontaneous emission A equal to 2×10^{-9} sec^{-1}, we estimate the stimulated transition rate to be 2 sec^{-1} if the maser is isotropic and 0.02 sec^{-1} if the maser radiates into a solid angle of 0.1 steradian. The net pumping rate is likely to be comparable with the infrared relaxation rate of the 6_{16} level, which should be somewhat faster than the spontaneous rate of 1 sec^{-1} because of stimulated infrared transitions. These estimates, based on the presently accepted upper limit on the sizes of some of the clouds, would thus indicate that the water maser may not be saturated.

The gain sensitivity of an unsaturated maser to changes in the population inversion is

$$\delta I_o / I_o = \delta(\alpha L)$$

For $(\alpha L) \approx 20$, as estimated above, a 1 percent change in (αL) causes a 20 percent change in intensity. The gain of a saturated maser is much less sensitive; the output intensity is directly proportional to the inverted population. If the large variations in the intensity are due to variations in maser gain, they are therefore easier to reconcile with the unsaturated maser.

If we assume amplification in an unsaturated H_2O maser with a gain constant of 19, the line width will be narrowed by a factor of about 4. The observed line width of about 40 khz then puts an upper limit on the kinetic temperature of the H_2O cloud of 1600°K. Turbulent line-broadening would lower this limit.

One possibility suggested by the apparent symmetrical pairing of some features in the W49 spectrum is that of orbital motions of clouds gravitationally bound to a central mass. For a discrete

cloud, a variation in radial velocity should be observed on a time scale determined by the orbital period, whereas, for a distributed cloud such as a ring, this would not be the case. The high-velocity features in the W49 spectrum suggest that a cloud with an orbital velocity greater than 100 km/sec would have an orbital period of a few days for a central mass of 1 solar mass and a few years for 100 solar masses. No appreciable shift in radial velocity of the features with high negative velocities in the W49 spectrum has been observed over 2 months' time, an indication that the central mass is probably larger than 10 solar masses if these features are associated with discrete clouds in orbital motion.

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Carbon Monoxide: Residence Time in the Atmosphere

Abstract. *A lower limit of 0.1 year for the residence time of carbon monoxide in the atmosphere is derived from radiocarbon measurements. The action of certain microorganisms and atmospheric photochemical reactions are possible mechanisms for the removal of carbon monoxide. This value can be compared with 2.7 years, a value deduced from estimated rates of carbon monoxide production and global measurements of atmospheric concentrations of carbon monoxide.*

Carbon monoxide was first reported in 1949 to be a minor constituent of the atmosphere by Migeotte (*1*), who assigned certain lines in the 4.7-μm region of the solar spectrum to atmospheric carbon monoxide. Bates and Witherspoon (*2*) estimated that the life of carbon monoxide in the atmosphere is less than 4 years. This estimate was based on a production rate for CO in 1951 of 8×10^{17} molecule cm^{-2} year^{-1} (1.9×10^{14} g year^{-1}) and a global concentration of 0.1 part per million (the lower of two values observed by Migeotte and Nevin in Switzerland) (*3*). Although these considerations suggested to them that CO was not accumulating in the atmosphere, they did not feel that the removal mechanism could be identified with certainty at that time.

Of the particular removal mechanisms for CO that they considered, the action of soil bacteria [in particular, *Bacillus oligocarbophilus* (*4*)] and the observations of Jones and Scott (*5*) were deemed the most significant, although the data were not suitable for quantitative analysis. (Jones and Scott, in studying the disappearance of carbon monoxide from sealed-off mine shafts after a coal mine fire, found that many materials from the mine or its vicinity were capable of removing carbon monoxide from air.) A more recent estimate of the lifetime of CO in the atmosphere has been made by Robinson and Robbins (*6*) who followed the approach of Bates and Witherspoon (*2*). They estimated the global CO production to be 2.1×10^{14} g year^{-1} in 1966 and the average atmospheric concentration to be 0.1 ppm (based on recent atmospheric measurements in remote areas). They derived a residence time for CO of 2.7 years in agreement with the previous estimate. I report here an independent method for estimating the lifetime of CO in the atmosphere that is related to the radiocarbon-dating method.

The radiocarbon-dating method (*7*) is based on the production of ^{14}C in the atmosphere by the reaction $^{14}N(n,p)^{14}C$ and the subsequent formation of radioactive CO_2. Measurements of the ^{14}C content of samples from the terrestrial biosphere and of contemporary shells, which were independent of latitude and longitude (*8*), indicated that this radioactive CO_2 is uniformly distributed over the surface of the earth. That ^{14}C was in a steady state in nature was indicated by the fact that the specific activity of ^{14}C measured in living things agreed with that calculated from the ^{14}C formation rate and the global carbon inventory (*9*). The success of this method for dating is well established (*10*).

However, in proposing the method, Libby assumed that the radioactive ^{14}C atoms would become CO_2 within a few hours after their formation. Subsequently, Pandow *et al.* (*11*) studied the chemical fate of ^{14}C produced in an atomic reactor in the presence of air and found that it was principally fixed as CO. They confirmed this result with ^{11}C produced in a heavy ion accelerator (*12*), where the possibility that the radioactive CO might have been produced in the reactor experiment by radiation decomposition was eliminated. Thus, if ^{14}C is initially fixed as CO, the rate of conversion of radioactive CO to radioactive CO_2 provides an independent method for determining the lifetime of CO in the atmosphere. MacKay *et al.* did, in fact, report measurements of the specific activity of atmospheric CO (*12*) from which the lifetime of CO can be estimated.

In collaboration with L. C. Matsch, they separated three samples of atmospheric CO from liquefied air produced by the Linde Air Products Company in their South Buffalo plant between 21 January and 7 March 1960. The sampling periods comprised 1 to 3 weeks and gave results for the ^{14}C specific activity of +9.3, −39.3, and −27.9 percent deviation relative to the 95 percent deviation of an oxalic acid sample (National Bureau of Standards) defined as zero age in 1955. The CO content of the air was approximately 0.3 ppm. Although they expected the specific activity of atmospheric CO to