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ASTRONOMY AND ASTROPHYSICS

# Searching for $^{16}O^{18}O$

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**Abstract.** After our first tentative detection of the molecular oxygen $^{16}O^{18}O$ isotopomer obtained during the winter '92/93 (Pagani et al. 1993), we attempted to confirm our results by making a deep search in other sources and in other positions in L134N. We have not been able to confirm the original detection in any other place in L134N but we obtain good upper limits towards NGC 2264(IRS2), TMC2, OMC3 and L134N (4', -1') and (5.2', -1) which give an $O_2$/CO ratio upper limit near 0.1 — the lowest yet obtained toward galactic sources — instead of 0.3–0.4 as predicted by gas phase chemical equilibrium models. As our sample consists of eight cold or lukewarm dark clouds, the low abundance of $O_2$ with respect to CO seems to be a general characteristic of dark clouds.

**Key words:** ISM: clouds; molecules; abundances – radio lines: ISM

## 1. Introduction

Oxygen is one of the most abundant elements in the universe and a key species in the interstellar chemistry. In the dense, cold molecular clouds, gas phase oxygen is predicted to be mostly in O, CO, $O_2$ and $H_2O$. However, until now only CO has been quantitatively measured, being the only one not severely blocked by the Earth's atmosphere. For these reasons, measurement of the molecular oxygen abundance is important to constrain the chemical models, especially for the C/O ratio in the gas phase. While detection of $^{16}O_2$ in our Galaxy requires the launch of balloon– or satellite–borne heterodyne receivers tuned to one of its lines, its $^{18}O$–substituted isotopomer can be observed from the ground, especially at 1.3 mm where its 234 GHz (N, J) = (2, 1) — (0, 1) line sits. Because its rotation lines are from magnetic dipole coupling (implying very small Einstein A–coefficients, Black & Smith 1984) and its abundance is low, this line has escaped detection for years for lack of sensitive–enough receivers and long–enough integration time. Even today, with the best performing SIS heterodyne receivers, the line seems almost impossible to detect.

There has been a renewed effort to detect $O_2$ in molecular clouds since telescopes have been equipped with sensitive SIS receivers (Combes et al. 1991., Fuente et al. 1991, Pagani et al. 1993, Combes & Wiklind 1995). The best 3 $\sigma$ upper limit in a single molecular cloud, $O_2/CO \leq 0.046$ has been obtained by the search at high redshift of the 424 GHz (N, J) = (2, 1) — (0, 1) $O_2$ line, redshifted to 252 GHz, in absorption in front of the B0218+357 quasar (Combes & Wiklind 1995). The background continuum source, B0218+357, allows one to observe a region as small as 5 pc in absorption. However, the lack of information about the absorbing molecular cloud could hide a clumpy structure or a local increase of the UV radiation field to explain this low ratio without introducing contradiction with the predictions of chemical models.

In our previous paper (Pagani et al. 1993 referred to as Paper I hereafter), we presented two tentative detections towards L134N and NGC 7538. The presence of three peaks in the L134N spectrum was unexpected because the source is a cold, dark cloud in which only a single component is seen in $C^{18}O$ and for which most of the exotic transitions seen in hot sources should not be seen here. For NGC 7538, we failed to reconfirm the detection during the same winter (1993) so we suspected it to be an artefact or due to a telescope pointing problem. The rms limits being already very low for these two sources (4 and 6 mK for 0.1 km/s velocity resolution for NGC 7538 and L134N, respectively), it would have taken an unrealistic amount of time to improve them significantly for L134N or to search around the central position for the possibly lost position in NGC 7538. So we preferred instead to explore new sources and to observe other positions in L134N during the next three winters. However, technical problems and a lack of good weather barely let us reach significant upper limits on a few other sources including three other positions in L134N.

## 2. Observations and results

The observations were made in several runs, spaced across three consecutive winters, at the POM–2 telescope from December

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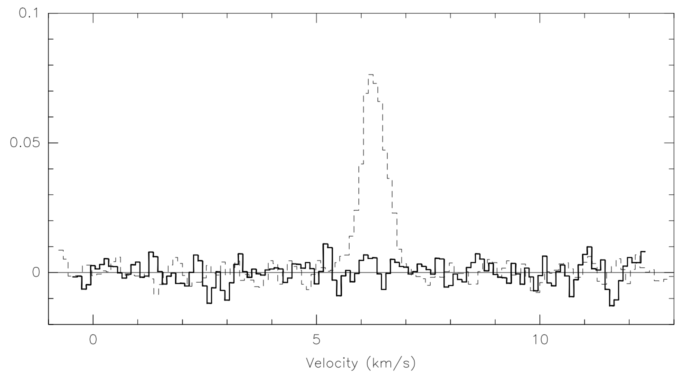


**Fig. 1.** TMC2 $C^{18}O$ (J:2–1) and $^{16}O^{18}O$ (N, J) = (2, 1) — (0, 1) spectra. The rms noise is 4.5 mK for 0.1 km/s resolution. Horizontal axis is the LSR Velocity (km $s^{-1}$) and vertical axis is the corrected radiation temperature $T_r^*$. The $C^{18}O$ line has been divided by 20.

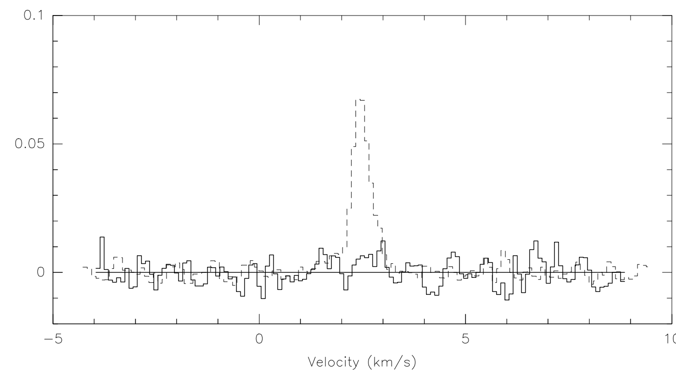


**Fig. 2.** L134N (2.5', -1') $C^{18}O$ (J:2–1) and $^{16}O^{18}O$ (N, J) = (2, 1) — (0, 1) spectra. The rms noise is 5.0 mK for 0.1 km/s resolution. The $C^{18}O$ line has been divided by 20.

1993 to January 1996 in the same way as during the first campaign (paper I). We were careful to check the receiver alignment and telescope pointing and we adopted a Double Position Switch technique (two reference positions in opposite directions) for NGC 7538 and DR21 to obtain a wider and flatter baseline and to cancel out residual ripples in the baseline. The receiver temperature was 70 K the first winter, 120 K the second winter and 70 K again in January 1996. Data reduction was conducted exactly in the same way as during the first campaign.

Getting useful 3 $\sigma$ upper limits has required between 150 and 200 hours of effective integration time per source. During the three winter campaigns, we were at the telescope a total of about 25 weeks with 28 % (7 weeks) of effective observing time (equivalent water vapor height $\leq$6 mm and absence of technical problems). Out of these seven weeks, we had only one week of very dry weather with an equivalent $H_2O$ height lower than 1 mm. If we had concentrated all this observing time on a single source, we would have reached a rms noise of only 2.3 mK.

The sources have been chosen to get their velocities Doppler–shifted enough to fall outside of the telluric line. Most of them are cold dark clouds ($T_{kin}\sim$10 K) because at higher temperature, a lot of other molecular lines are excited and some of them could fall near the frequency of $^{16}O^{18}O$. As cold dark clouds often have low $C^{18}O$ column densities, we have also selected three lukewarm ($\sim$ 20 K) sources with significantly higher $C^{18}O$ column densities: DR21, NGC 2264(IRS2) and OMC3. The inclusion of these sources is a reasonable trade–off of the available observing time since the $^{16}O^{18}O$ line is predicted to decrease by less than 20 % in intensity from 10 to 20 K while the $C^{18}O$ column density in these sources is 2 to 3 times higher than in cold sources like TMC2 or L134N. As the photodissociation rate of molecular oxygen is an order of magnitude higher than that of CO, all the sources have been chosen without any intense UV radiation field on the scale of our beamsize.

The three positions in L134N were chosen to be the $C^{18}O$ (J:1–0) peak (4', -1'), the $C^{18}O$ (J:2–1) peak (5.2', -1') (Pagani & Pardo 1996) and the SO and $SO_2$ peak (4.-1', 8.-0') (Swade 1989) all measured as offsets from the nominal center (Table 1).

The results of our observations are summarized in Table 1 and the spectra of TMC2, L134N (2.5', -1'), NGC 2264(IRS2) and DR21 are displayed in Figs. 1 to 4. The TMC2 spectrum (Fig. 1) which has the lowest rms noise (4.5 mK for 0.1 km/s velocity resolution) shows no line artefact and a noise close to the theoretical prediction ($\Delta T=\sqrt{2}\, T_{sys}/\sqrt{B\tau} \sim 5.5$ mK). A similar result is found for the L134N (2.5', -1') spectrum (Fig. 2) and NGC 2264(IRS2) spectrum (Fig. 3) which has the lowest $O_2$/CO limit ever obtained for a galactic cloud. DR21 (Fig. 4) shows that even with a 36 MHz bandwidth the noise is still normal. $C^{18}O$ column densities are obtained using our AT&T Bell Labs $C^{18}O$ (J:1–0) and POM–2 $C^{18}O$ (J:2–1) observations. Upper limits are given as 3 times the rms deviation of a channel the width of the $C^{18}O$ line.

To estimate the $^{16}O^{18}O$ column density limits, we assume that the lines are optically thin and that the levels are thermalized. The column density is given by:

$$N\left(^{16}O^{18}O\right) = \frac{8\ 10^{15} \int \Delta T_r^* \delta v}{f\,(T_{kin})\, e^{-h\nu/kT_{kin}}} \quad (1)$$

where $f(T_{kin})$ is the fraction of the population in the (N, J) = (0, 1) level at the temperature $T_{kin}$, $\Delta T_r^*$ is the antenna excess temperature over the 2.7 K background. $T_{kin}$ is assumed to be 10 K for cold sources (B5, TMC2, L134N and B335), 20 K for OMC3, NGC 2264(IRS2) and DR21, and 40 K for NGC 7538.

## 3. Discussion

To interpret our limits on the $^{16}O^{18}O$ ratio we need theoretical models of carbon and oxygen chemistry, preferably including isotopic exchange reactions. Many groups have developed models that calculate the $O_2$/CO ratio in interstellar clouds under a variety of conditions (see for a review van Dishoeck 1988), but only two include all the carbon and oxygen isotopes explicitly (Langer et al. 1984, and Maréchal et al. 1996). Here we use the Maréchal et al. model to interpret our data as it uses a more recent chemical reaction data base, and considers both photodis-

**Table 1.** Source list, observational results, estimates of $C^{18}O$ and $^{16}O^{18}O$ column densities, $O_2$/CO ratio and C/O elemental abundance ratio

| Source | R.A<br>h min s | Dec<br>deg min s | $C^{18}O$ $\delta$v<br>km s$^{-1}$ | $^{16}O^{18}O$ $\int \Delta T_r^* \delta v$<br>mK km s$^{-1}$ | N($C^{18}O$)<br>cm$^{-2}$ | N($^{16}O^{18}O$)<br>cm$^{-2}$ | $O_2$/CO | C/O |
|---|---|---|---|---|---|---|---|---|
| B5 | 3 44 29 | +32 44 30 | 0.8 | $\leq$8.5 | 2.1 $10^{15}$ | $\leq$1.1 $10^{15}$ | $\leq$0.25 | $\geq$0.43 |
| TMC2 | 4 29 43 | +24 16 55 | 0.8 | $\leq$3.8 | 2.1 $10^{15}$ | $\leq$4.9 $10^{14}$ | $\leq$0.11 | $\geq$0.57 |
| OMC3 | 5 42 47 | -5 01 28 | 1.3 | $\leq$7.0 | 3.8 $10^{15}$ | $\leq$1.0 $10^{15}$ | $\leq$0.13 | $\geq$0.55 |
| NGC 2264(IRS2) | 6 38 16 | +9 38 14 | 3.0 | $\leq$8.7 | 7.0 $10^{15}$ | $\leq$1.2 $10^{15}$ | $\leq$0.086 | $\geq$0.60 |
| L134N | 15 51 30 | -2 43 30$^a$ | | | | | | |
| (2.5', -1') | | | 0.5 | $\leq$3.4 | 1.2 $10^{15}$ | $\leq$4.4 $10^{14}$ | $\leq$0.16 | $\geq$0.51 |
| (4', -1') | | | 0.5 | $\leq$5.8 | 2.5 $10^{15}$ | $\leq$7.5 $10^{14}$ | $\leq$0.15 | $\geq$0.52 |
| (-1.4', -0.8') | | | 0.5 | $\leq$5.3 | 1.1 $10^{15}$ | $\leq$6.8 $10^{14}$ | $\leq$0.26 | $\geq$0.42 |
| B335 | 19 34 34 | +7 27 00 | 0.38 | $\leq$5.0 | 3.0 $10^{14}$ | $\leq$6.5 $10^{14}$ | $\leq$0.62 | $\geq$0.26 |
| DR21 | 20 37 14 | +42 08 55 | 3.0 | $\leq$17.7 | 5.3 $10^{15}$ | $\leq$2.6 $10^{15}$ | $\leq$0.23 | $\geq$0.45 |
| NGC 7538 | 23 11 36 | +61 11 47 | 4.4 | $\leq$23.4 | 3.5 $10^{15}$ | $\leq$5.1 $10^{15}$ | $\leq$0.72 | $\geq$0.24 |

$^a$ central position (0', 0')

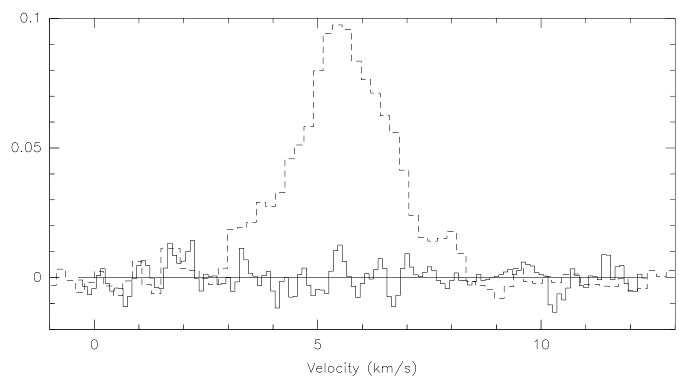


**Fig. 3.** NGC 2264(IRS2) $C^{18}O$ (J:2–1) and $^{16}O^{18}O$ (N, J) = (2, 1) — (0, 1) spectra. The rms noise is 5.3 mK for 0.1 km/s resolution. The $C^{18}O$ line has been divided by 30.

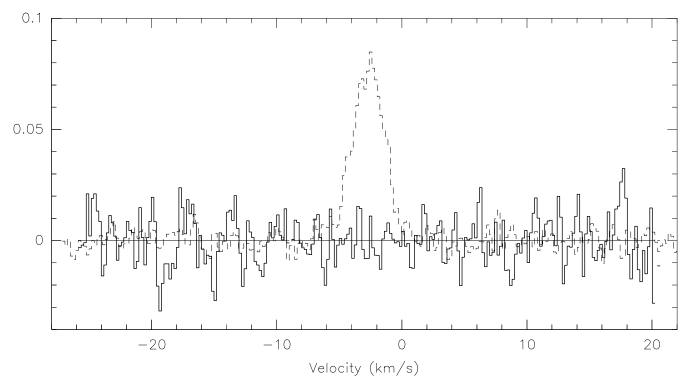


**Fig. 4.** DR21 $C^{18}O$ (J:2–1) and $^{16}O^{18}O$ (N, J) = (2, 1) — (0, 1) spectra. The rms noise is 11 mK for 0.2 km/s resolution. The $C^{18}O$ line has been divided by 40.

sociated and shielded regions. The time dependent models of Langer et al. were restricted to shielded cores.

The model of Maréchal et al. allows us to estimate the isotopic ratios needed to convert the $^{16}O^{18}O/C^{18}O$ ratio into the more useful $O_2$/CO ratio. The results are close to those obtained by assuming $^{16}O^{16}O/^{16}O^{18}O$ = 250 and $C^{16}O/C^{18}O$ = 500 except for low $C^{18}O$ column densities as in B335 where $C^{18}O$ is less self–shielded than CO in which case the $C^{16}O/C^{18}O$ ratio is predicted to reach 870. These model results are similar to those found by Langer et al. (1984) for isotopic ratios in shielded regions. The lower limits upon the C/O ratio corresponding to those column densities are deduced from the same model.

Steady state models or time-dependent models reaching chemical equilibrium ($\sim 10^7$ yr) predict an $O_2$/CO ratio quite independent of density and kinetic temperature and equal to 0.3–0.4 in a quiescent dark molecular cloud if we assume a C/O elemental abundance ratio of 0.44 (cosmic value) (e.g. Leung et al. 1984, Pineau des Forêts et al. 1991, Maréchal et al. 1996). Our observational results imply that the $O_2$ abundance in dark clouds is lower than this value by a factor $\geq$ 3. If we assume that the chemical models are accurate enough concerning the chemistry of the oxygen–bearing molecules, then the C/O elemental abundance ratio could be larger than the cosmic value by 35 % or more to match our results. A way to get the C/O ratio higher than the cosmic value is to assume a selective O–depletion on grains, more particularly on grain mantles in form of $H_2O$, CO, $O_2$ and $CO_2$ ices (Langer et al. 1984, Blake et al. 1987). Due to the uncertainties about the chemical reactions on grain surfaces and the efficiency of desorption mechanism (Hasegawa & Herbst 1993, Bergin et al. 1995), quantitative estimates cannot be easily used to constrain the gas phase C/O ratio.

One could interpret those low $O_2$/CO ratios without need to change the C/O elemental abundance ratio. $O_2$ is mainly destroyed by collisions with $C^+$, $He^+$ and $H^+$ in the dark clouds and thus an underestimate of the ionization of the clouds by the chemical models could explain the deviation from the observations. Le Bourlot et al. (1995) have obtained models with a high ionization phase in which $O_2$ fractional abundance is lower than usual by a factor of 100 without changing significantly the CO abundance. In the range of densities prevailing in cold dark clouds, the two ionization phases (low and high) could be obtained by these models and observations are needed to distinguish between them. Similarly, one could ask oneself about the way to produce $O_2$. The reaction rate of the main production way of $O_2$, O+OH→$O_2$+H, is not well known but, as Black & Smith (1984) already showed, we found that its increase or decrease is compensated by the inverse behaviour of OH abundance in dark clouds (Maréchal et al. 1996) and eventually, the $O_2$ abundance remains constant. Thus, the uncertainty on this reaction rate cannot be involved to explain the low $O_2$ fractional abundances deduced from the observations. Another possibility can also be investigated from the Sternberg and Dalgarno work (1995). These authors have modeled the chemistry of regions with high UV radiation fields and very high densities, including the effects of cosmic ray induced photodissociation. The latter effect can reduce the abundance of $O_2$ even in shielded regions and they find a value for the fractional abundance, X($O_2$), about $10^{-6}$ at n($H_2$) = $10^6$ cm$^{-3}$, below the upper bounds set by our observations. However, model calculations at lower densities of the cores observed here (about a few $10^4$ cm$^{-3}$) yield higher values of X($O_2$) greater than $10^{-5}$ (Bergin and Langer, private communication). Hence we believe that our limits on $O_2$ must be explained by some other effects.

Finally, time dependent models omitting depletion predict that the $O_2$/CO ratio increases with time reaching a constant value after a duration of 10 Myr (Langer et al. 1984, Bergin et al. 1995). The increase of the $O_2$/CO ratio is about a factor of 25 between $10^5$ years and its constant limit. The underabundance of $O_2$ deduced from the observations could be a signature of the age of the clouds but cold dark clouds seem to be near virial equilibrium. On the other hand, mixing between the core and the photo–dissociated edges of the clouds (Pineau des Forêts et al. 1992, Xie et al. 1995) gives similar low $O_2$/CO ratios as those found at early times in time dependent models. The same results are also obtained by assuming the clumping of the clouds, in which case UV radiations can penetrate deeper inside the clouds, increasing the direct UV destruction of $O_2$ and also its destruction via an increased abundance of $C^+$.

## 4. Conclusion

We have not confirmed our first tentative detection of $^{16}O^{18}O$ in L134N at any of three other positions in this source, and thus have to consider this detection very cautiously. In addition we have not detected $^{16}O^{18}O$ in any of seven other clouds.

The $O_2$/CO upper limits we obtain begin to be really in conflict with gas phase chemical equilibrium model predictions assuming standard cosmic abundances for half of the sources we observed: NGC 2264(IRS2), TMC2, OMC3, L134N (5.2', -1') and L134N (4', -1'). They are the lowest limits yet obtained towards galactic sources. The high proportion of clouds having a very low $O_2$/CO ratio seems to suggest that a general phenomenum reduces the $O_2$ abundance in dark clouds with respect to the chemical model predictions and not that the results could be due to the observations of an atypical cloud as could be the case for the Combes & Wiklind (1995) upper limit found towards a single extragalactic molecular cloud. To interpret such a low $O_2$ abundance, we have to assume either a higher than expected ionization of the clouds, due to processes such as mixing or clumpiness, or a higher C/O ratio than the cosmic value which could be produced by the depletion of oxygen–bearing species on grain mantles. Ehrenfreund et al. (1992) suggested the possibility of detecting solid $O_2$ and other components of interstellar mantle ices with ISO satellite and these observations could be useful to constrain the gas phase elemental abundances.

It appears that we must wait for the SWAS, ODIN satellite or the PIROG and PRONAOS balloon results to make any further progress on the oxygen problem in our Galaxy.

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