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## The Dark Neutral Medium is (Mostly) Molecular Hydrogen

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#### ABSTRACT

*Context*. More gas is sometimes inferred in molecular cloud complexes than is represented in HI or CO emission, and this is called dark neutral medium (DNM).

*Aims.* Our aim is to extend a study of DNM along 13 directions in the outskirts of Chamaeleon by determining the atomic or molecular character of the DNM along a larger sample of sightlines.

*Methods.* We acquired ALMA ground rotational state absorption profiles of HCO<sup>+</sup> and other molecules toward 33 compact extragalactic continuum background sources seen toward the Galactic anticenter, deriving  $N(H_2) = N(HCO^+)/3 \times 10^{-9}$  as before. We observed J=1-0 CO emission with the IRAM 30m telescope in directions where HCO<sup>+</sup> was newly detected.

*Results.* HCO<sup>+</sup> absorption was detected in 28 of 33 new directions and CO emission along 19 of those 28. The five sightlines lacking detectable HCO<sup>+</sup> have three times lower  $\langle E_{B-V} \rangle$  and  $\langle N(DNM) \rangle$ . Binned in  $E_{B-V}$ ,  $N(H_2)$  and N(DNM) are strongly correlated and vary by factors of 50-100 over the observed range  $E_{B-V} \approx 0.05 - 1$  mag, while N(HI) varies by factors of only 2-3. On average N(DNM) and N(H<sub>2</sub>) are well matched, and detecting HCO<sup>+</sup> absorption adds little to no H<sub>2</sub> in excess of the previously inferred DNM. There are five cases where  $2N(H_2) < N(DNM)/2$  indicates saturation of the HI emission profile. For sightlines with  $W_{CO} \ge 1$  K-km s<sup>-1</sup>, the CO-H<sub>2</sub> conversion factor  $N(H_2)/W_{CO} = 2 - 3 \times 10^{20}$  cm<sup>-2</sup>/(1 K-km s<sup>-1</sup>) is higher than is derived from studies of resolved clouds in  $\gamma$ -rays.

*Conclusions.* Our work sampled primarily atomic gas with a mean H<sub>2</sub> fraction  $\approx 1/3$ , but the DNM is almost entirely molecular. CO fulfills its role as an H<sub>2</sub> tracer when its emission is strong, but large-scale CO surveys are not sensitive to H<sub>2</sub> columns associated with typical values N(DNM) =  $2 - 6 \times 10^{20}$  cm<sup>-2</sup>. Lower X<sub>CO</sub> values from  $\gamma$ -ray studies arise in part from different definitions and usage. Sightlines with W<sub>CO</sub>  $\geq 1$  K-km s<sup>-1</sup> represent 2/3 of the H<sub>2</sub> detected in HCO<sup>+</sup> and detecting 90% of the H<sub>2</sub> would require detecting CO at levels W<sub>CO</sub>  $\approx 0.2$ -0.3 K-km s<sup>-1</sup>.

Key words. interstellar medium – abundances

### 1. Introduction

Between the atomic and molecular interstellar gases there is a transition that is imperfectly traced in  $\lambda 21$ cm HI and/or  $\lambda 2.6$ mm CO emission. Gas that is not well represented in one or both tracers is described as dark neutral medium (DNM) (Grenier et al. 2005; Planck Collaboration et al. 2015; Remy et al. 2017, 2018), and the atomic or molecular character of the DNM can only be decided by appealing to other information.

In the outskirts of Chamaeleon, we used millimeter-wave  $HCO^+$  absorption to show that the DNM was almost exclusively molecular, even while the gas as a whole was mostly atomic (Liszt et al. 2018).  $HCO^+$  was detected along all 13 sampled sightlines, of which 12 lacked detectable CO emission at a level of 1.5 K-km s<sup>-1</sup>. The amount of inferred H<sub>2</sub> was comparable to that of the DNM (confirming the amount of the detected DNM), and only for three sightlines did it seem likely that saturation of the HI emission profile could account for much of the DNM. Subsequent detections of CO absorption along six directions by Liszt et al. (2019) showed that the lack of CO emission was due to low CO column densities. The linewidth of CO absorption was shown to be smaller than that of its parent molecule  $HCO^+$ , illustrating the complex chemical nature of the CO formation process in diffuse gas.

In this work we extend the analysis of the composition of DNM to 33 sightlines in directions toward the Galactic anticenter (Remy et al. 2017, 2018) and consider the larger sample of 46 sightlines. The present study employs a relatively small number of diffuse ( $A_V \le 1$  mag) and translucent ( $A_V \le 3$  mag) sightlines where background sources are serendipitously available, and is not a revision of the earlier study, whose derived DNM column densities and other results are assumed here. Rather, we use the newly derived  $H_2$  abundances, independent of the presence of CO emission, to consider such topics as the atomic or molecular character of the DNM, the degree to which the L21 HI profile represents N(HI) and the influence of the CO-H<sub>2</sub> conversion factor on the DNM determination.

Section 2 summarizes the observational material considered here and Section 3 compares the prior results for DNM with atomic hydrogen measured in the  $\pounds$ 21 emission and H<sub>2</sub> as sampled by its proxy 89 GHz absorption of HCO<sup>+</sup>. The role of CO emission data is explored in Section 4 where the CO-H<sub>2</sub> conversion factor is derived. Section 5 presents a summary, with conclusions and a discussion of later work.

### 2. Observations and data reduction

The methods of work were described in Liszt et al. (2018) and are not repeated in full detail here. From the existing DNM

analyses of Planck Collaboration et al. (2015) and Remy et al. (2017, 2018) we take N(DNM) and Ł21 HI column densities N(HI) and N(HI)<sub>cl</sub> representing respectively the line profile integral integrated over all velocities and the column density that is associated with the cloud and/or kinematic features hosting the neutral gas and DNM, as derived by a decomposition that accounts for the compound nature of HI emission profiles from individual clouds.

To the existing analyses we add ALMA observations of absorption from HCO<sup>+</sup> and profiles of  $\lambda 2.6$ mm CO emission from the IRAM 30m telescope, as described below.

#### 2.1. Millimeter-wave absorption

The new absorption data discussed here are spectra of HCO<sup>+</sup> in 33 directions along Galactic anticenter sightlines toward ALMA phase calibrators as given in the Tables here and projected on sky maps in Figure 1. As before, the HCO<sup>+</sup> spectra were acquired with a spectral resolution of 0.205  $\text{km}\,\text{s}^{-1}$  sampled at  $0.102 \text{ km s}^{-1}$  intervals without the so-called spectral averaging that bins data. We also acquired spectra of HCN, C<sub>2</sub>H, HCO, and several isotopologs. Counts of detections are shown in Figure 2, but results for species other than HCO<sup>+</sup> will be discussed in detail in later work. Statistics of the HCO<sup>+</sup> detections are shown in Figure A.1. Spectra in the 28 directions with detectable HCO<sup>+</sup> absorption are shown in Figure B.1.

#### 2.2. IRAM 30m CO emission

We took frequency-switched CO emission profiles at the IRAM 30m telescope in August 2019 toward the 28 anticenter sightlines with detected HCO<sup>+</sup> absorption. The CO spectra are shown in Figure B.1. These CO data were taken as five-point maps toward the target and displaced by 2 HPBW (2x22") in the four cardinal directions, using a pointing pattern that was designed to detect HCO<sup>+</sup> and HCN emission in the presence of spectral line absorption in the target direction. For the HCO<sup>+</sup> emission that will be discussed in subsequent work, the  $\mathrm{HCO}^+$  emission profile is formed by averaging spectra along the four outlying directions. For CO the present results use the average of all five pointings because contamination by absorption is not measurable given the strength of the emission and/or the continuum. The results are presented on the main beam antenna temperature scale that is native to the 30m telescope.

#### 2.3. Conversion from integrated HCO<sup>+</sup> absorption to $N(H_2)$

The suitability of HCO<sup>+</sup> as a proxy for H<sub>2</sub> in diffuse molecular gas was explored extensively in Liszt & Gerin (2023) (hereafter Paper 1). As in our earlier work, we use  $N(H_2) = N(HCO^+)/3 \times$  $10^{-9}$  and N(HCO<sup>+</sup>) =  $1.10 \times 10^{12} \Upsilon_{HCO^+}$  where  $\Upsilon_{HCO^+}$  is the integrated  $HCO^+$  optical depth expressed in km s<sup>-1</sup>.

#### 2.4. Reddening and dust optical depth

The 6' resolution dust-emission maps scaled to optical reddening  $E_{B-V}$  by Schlegel et al. (1998) are cited here. Those reddening values can be converted to Planck 353 GHz dust optical depth using the relationship established by Planck Collaboration et al. (2014) between the 353 GHz dust optical depth and the  $E_{B-V}$ values of Schlegel et al. (1998),  $E_{B-V}/\tau_{353} = (1.49 \pm 0.03) \times 10^4$ mag.

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#### 2.5. Conventions

Velocities presented with the spectra are taken with respect to the kinematic definition of the Local Standard of Rest. N(H) is the column density of H nuclei detected in neutral atomic and molecular form,  $N(H) = N(HI)+2N(H_2)$ , and the molecular hydrogen and DNM fractions 2N(H<sub>2</sub>)/N(H) and N(DNM)/N(H) are respectively represented by  $f_{H_2}$  and  $f_{DNM}.$  The integrated absorption of the HCO<sup>+</sup> profile in units of km s^{-1} is denoted by  $\Upsilon_{HCO^+}$ and similarly for other species. The integrated emission of the J=1-0 CO line is denoted by  $W_{CO}$  with units of K-km s<sup>-1</sup> and the CO-H<sub>2</sub> conversion factor  $N(H_2)/W_{CO}$  is denoted by  $X_{CO}$ . Where reference is made to a typical Galactic or standard CO-H<sub>2</sub> conversion factor, the value N(H<sub>2</sub>)/W<sub>CO</sub> =  $2 \times 10^{20}$  cm<sup>-2</sup>/(1 Kkm s<sup>-1</sup>) should be understood, as summarized in Table E.1 of Remy et al. (2017).

#### 2.6. Overview

Observational results of importance to this work are given in Table 1 and Table 2, and summaries of mean properties of various subsamples are presented in Table 3. Some statistics of the continuum targets and noise in the detections of HCO<sup>+</sup> absorption are discussed in Appendix A and the CO emission and HCO<sup>+</sup> absorption line profiles are shown in Figure B.1 and discussed in Appendix B.

## 3. HI, DNM, and H<sub>2</sub> sampled in HCO<sup>+</sup> absorption

Figure 1 shows the new Galactic anticenter sightlines projected on maps of  $E_{B-V}$  and N(DNM). The region at  $b < -30^{\circ}$  is described by Remy et al. (2017) as Cetus, and that at  $b > -22^{\circ}$  as Taurus-Main. California lies to the north around  $b = -10^{\circ}$  and Perseus is near  $(1,b) = 160^{\circ}, -20^{\circ}$ . Taurus-North overlays most of the map region. The individual subregions are not discussed here owing to the sparse sampling.

Counts of sources and detected molecular tracers binned in 0.05 mag intervals of reddening are shown in Figure 2. HCO<sup>+</sup> is the molecular tracer detected most commonly (28 of 33 anticenter sightlines and 41 of 46 overall), followed by  $C_2H$  (26 total) and HCN (21 total). The properties of molecular absorption line tracers other than HCO<sup>+</sup> will be discussed in later work.

The Chamaeleon and Galactic anticenter subsamples have the same mode at  $E_{B-V} = 0.25 - 0.3$  mag in Figure 2, but the anticenter subsample has a high-reddening tail at  $E_{B-V} > 0.5$  mag that is absent in Chamaeleon. The Galactic anticenter sample has higher mean extinction and molecular fraction. Overall (see Table 3) the anticenter sources have the following characteristics:

- 33% higher  $\langle E_{B-V} \rangle = 0.36$  vs 0.27 mag;
- Same  $\langle N(HI) \rangle = 1.3 \times 10^{21} \text{ cm}^{-2}$  and  $f_{DNM} = 0.12$ ; 80% higher  $\langle N(HI)_{cl} \rangle = 1.12 \text{ vs } 0.62 \times 10^{21} \text{ cm}^{-2}$ ;
- 2 times higher  $\langle 2N(H_2) \rangle = 0.80 \text{ vs } 0.39 \times 10^{20} \text{ cm}^{-2}$ ;
- 65% higher  $f_{H_2} = 0.38$  vs 0.23; \_
- \_ Higher fraction of detected CO, 9/33 (18/33 in the new work) vs 1/13;
- Higher incidence of nondetection of HCO<sup>+</sup>, 5/33 vs 0/13, in some cases due to low flux, see Figure A.1.

The DNM fraction is nearly the same in the two samples (0.12-0.13) and much smaller (0.06) in both subsamples when CO emission was detected at the level of 1 K-km s<sup>-1</sup>. The DNM fraction is also noticeably smaller when molecular gas sampled in HCO<sup>+</sup> absorption is absent, and similarly at  $E_{B-V} < 0.2$  mag. The strongest variations in f<sub>DNM</sub> in Table 3 arise in selections



Fig. 1. Sky maps of the observed sightlines. Left: Positions of the background sources observed here are projected against a map of  $E_{B-V}$  from the work of Schlegel et al. (1998). The sightlines lacking detected HCO<sup>+</sup> absorption (largely due to weak continuum) are indicated with smaller symbols. Right: Same as left, but shown against a map of N(DNM) from Remy et al. (2017, 2018).

based on the strength of CO emission and are discussed in Section 4.

#### 3.1. Quantitative summary results

Figure 3 shows an overview of trends in means of N(HI), N(DNM), and  $N(H_2)$  using data binned in 0.05 mag intervals of reddening as in Figure 2, plotted horizontally at the mean  $E_{B-V}$ in every bin. The counts in each bin are shown and the bins at  $E_{B-V} > 0.5$  mag are occupied by only one or two sightlines. The association of DNM with molecular gas is unmistakable.

The mean HI column density varies by only a factor of 3 over a wider range  $E_{B-V} = 0.09 - 1$  mag. By contrast, means of N(NDM) and N(H<sub>2</sub>) vary by factors of 50-100. The values of  $\langle N(DNM) \rangle$  and  $\langle N(H_2) \rangle$  are comparable and increase steadily for  $E_{B-V} \leq 0.7$  mag, beyond which  $\langle N(DNM) \rangle$  either fails to increase or falls: the CO emission tracer used to derive N(DNM) was used effectively to account for H nuclei in H<sub>2</sub>. The cloudassociated mean N(HI)<sub>cl</sub> declines at  $E_{B-V} \ge 0.7$  mag as H<sub>2</sub> sequesters H nuclei. The value of  $\langle N(H_2) \rangle$  increases up to the bins at highest  $E_{B-V}$  where  $\langle N(H_2) \rangle \approx \langle N(HI) \rangle \approx 2 - 3 \langle N(HI)_{cl} \rangle$ and whole sightlines are dominated by molecular gas.

The correlation between DNM and H<sub>2</sub> is remarkable considering the vastly different scales on which  $E_{B-V}$ , N(HI), and N(NDM) are measured (6-20'), as contrasted with N(H<sub>2</sub>)  $\propto$  $\Upsilon_{HCO^+}$  that is derived on submilliarcsecond scales from absorption against scatter-broadened compact millimeter-wave ALMA calibrator continuum sources. The same kind of correlation of tracers observed on different angular scales occurs in the correlation of integrated  $\lambda 21$  cm HI optical depth with  $E_{B-V}$  (Liszt 2021).

When  $HCO^+$  is not detected (5/46 directions) the following characteristics are found compared to sightlines with detected HCO<sup>+</sup>:

- 3 times lower  $\langle E_{B-V} \rangle = 0.13$  mag;
- 1.3 times higher  $\langle N(H) \rangle / \langle E_{B-V} \rangle = 8 \times 10^{21} \text{ cm}^{-2} \text{ mag}^{-1}$ ; 3.5 times lower  $\langle N(DNM) \rangle = 8 \times 10^{19} \text{ cm}^{-2}$ ;



Fig. 2. Counts of sightlines and detected tracers binned in 0.05 intervals of reddening, E<sub>B-V</sub>. The histogram of all sources is overlaid (dashed red) in the other panels.

- 2 times lower  $< f_{DNM} > = 0.07;$
- no DNM in 3/5 cases vs. 15/46 overall.

Instead, using  $3\sigma$  upper limits for N(H<sub>2</sub>) we find:

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- >5 times lower <2N(H<sub>2</sub>)>
$$\leq$$
 1.5 × 10<sup>20</sup> cm<sup>-2</sup> and

>2.5 times lower  $< f_{H_2} > \le 0.14$ 

The ensemble of sightlines with N(DNM)  $< 5 \times 10^{19} \text{ cm}^{-2}$ does not differ by more than 10-20% from other sightlines in any mean property not involving N(DNM). The variations in N(DNM) and N(H<sub>2</sub>) stand in great contrast with N(HI) that increases only by a factor of 3 across the figure. The rise in  $N(H_2)$ accounts for the failure of N(HI) to increase at high  $E_{B-V}$ , but this could in principle also be explained by increasing saturation of the HI emission. As noted in Planck Collaboration et al. (2015) and further refined by Remy et al. (2018), the global DNM analysis finds the best fit with the optically thin estimate of N(HI), implicitly leaving a bigger overall role for molecular gas than for saturation of the HI profile.

#### 3.2. DNM and HI

Shown in Figure 4 is the variation of <N(DNM)> with <N(HI)> and <N(HI)<sub>cl</sub>>. Chamaeleon differs from the Galactic anticenter in having more extraneous atomic gas along the line of sight and smaller values of cloud-associated gas. The cloud-associated HI in Chamaeleon is clustered at the low end of the range, even though the samples are evenly matched in the span of their total HI column density and have the same mean N(HI) in Table 3.

Sightlines lacking DNM are present at all N(HI), but N(DNM) is uniformly small along sightlines at the highest column densities N(HI)  $\gtrsim 2.3\times 10^{21}~cm^{-2}$  where CO emission is more commonly observed. The sources with N(DNM) > 6  $\times$  10^{20} cm^{-2} and high N(DNM)/N(HI) ratios are perhaps the most obvious candidates for hosting significant quantities of DNM in atomic form as the result of saturation of the HI profile, but only two of these actually have small contributions from N(H<sub>2</sub>). Assessing the DNM contribution arising from possible saturation of the HI profile is discussed in Section 3.3 where it is seen that the strongest cases of atomic DNM actually have more modest N(DNM).

#### 3.3. DNM, $E_{B-V}$ , and $N(H_2)$

As with HI, sightlines lacking DNM are present over the full range of  $E_{B-V}$  in Figure 5, top, but they are predominant at smaller N(H<sub>2</sub>). Sightlines with 2N(H<sub>2</sub>)  $\leq 10^{20}$  cm<sup>-2</sup> lack DNM. Along sightlines with column densities too low to host molecular gas, the atomic gas is well represented by the optically thin estimates of N(HI) used in the DNM analysis.

Care must be taken in interpreting the relationship of DNM and N(H<sub>2</sub>) in Figure 5 because the DNM in part represents material that is missing in the CO emission tracer while some of the H<sub>2</sub> traced by HCO<sup>+</sup> is actually visible in CO emission. To minimize this cross-contamination, Figure 6 shows a plot of N(DNM) vs. N(H<sub>2</sub>) for sightlines lacking a detection of CO, using the more sensitive IRAM data for the Galactic anticenter subsample. Included are all but one (12/13) of the Chamaeleon sightlines and 60% of those toward the anticenter.

Sightlines lacking DNM in the absence of CO emission in Figure 6 are almost entirely confined at  $2N(H_2) \leq 2 \times 10^{20} \text{ cm}^{-2}$ , so the detections of HCO<sup>+</sup> absorption do not imply much additional gas along sightlines where DNM was not found. Missing from Figure 6 are sightlines with  $2N(H_2) \gtrsim 10^{21} \text{ cm}^{-2}$  corresponding to  $W_{CO} = 2.5 \text{ K-km s}^{-1}$  for the Galactic CO-H<sub>2</sub> conversion factor.

There are two or three cases in each subsample where  $N(DNM)/2N(H_2) \gtrsim 2$  and saturation of the  $\pounds 21$  emission profile may be important. Most of these sightlines have modest values N(DNM)  $\approx 4 - 5 \times 10^{20}$  cm<sup>-2</sup> and are not the sightlines with high N(DNM) that were singled out for discussion in Section 3.2 when comparing N(DNM) and N(HI).

There are also two or three sightlines at  $N(H_2) > 6 \times$  $10^{20}~\text{cm}^{-2}$  where N(DNM)/2N(H\_2)  $\stackrel{\scriptstyle <}{\scriptstyle \sim}$  2. Overall <2N(H\_2)> = 1.3<N(DNM)> for sources lacking CO emission, so H<sub>2</sub> accounts for DNM without adding extra "dark" gas, and the DNM is mostly molecular. As before in Chamaeleon, the DNM is largely molecular, even if the medium overall is predominantly atomic.

## 4. The role of CO emission

The CO emission that was detected in previous studies used to determine N(DNM) along our sightlines was well above 1 Kkm s<sup>-1</sup> in every case (Table 2), with  $\langle W_{CO} \rangle = 4.9$ K-km s<sup>-1</sup> (Table 3). This is very nearly the same mean as for the sample of sightlines with  $W_{CO} \ge 1$ K-km s<sup>-1</sup> observed at the IRAM 30m, 5.5 K-km s<sup>-1</sup> (see Table 2). Compared to the overall average, the sightlines with  $\langle W_{CO} \rangle \ge 1 \text{ K-km s}^{-1}$  have the following characteristics:

- 50-100% higher  $\langle E_{B-V} \rangle = 0.5 0.7$  mag;
- 3-4 times higher  $<2N(H_2)> = 2 3 \times 10^{21} \text{ cm}^{-2}$ ; 2 times higher  $<f_{H_2}> = 0.6$ ;
- 2 times lower  $\langle f_{DNM} \rangle = 0.06$ .

The 50% smaller DNM fractions  $<\!f_{DNM}\!>$  = 0.06 along samples of sightlines with  $<\!W_{CO}\!>$   $\geq$   $1K\text{-}km\,s^{-1}$  indicate that CO emission is doing a good job of tracing  $H_2$  in gas where the molecular fraction is high (Liszt 2017), CO emission is strong, and the cloud-associated HI fraction declines (Figure 2). However, CO emission in general represents a small portion of the total molecular gas present along the sightlines in this work. For instance,  $\langle 2N(H_2) \rangle = 7 \times 10^{20} \text{ cm}^{-2}$  along 46 sightlines, while  $\langle W_{CO} \rangle \approx 5 \text{ K-km s}^{-1}$  along the 6-10 sightlines where  $\langle W_{CO} \rangle \geq 1 \text{ K-km s}^{-1}$  (Table 3). For a typical Galactic CO-H<sub>2</sub> conversion factor, the summed molecular gas column represented in HCO<sup>+</sup> is three to four times that inferred from the summed CO emission. Most of the molecular gas in our sample was hidden in the DNM prior to our work and is still only revealed by the HCO<sup>+</sup> absorption, even with more sensitve CO observations. The IRAM 30m detections with  $\langle W_{CO} \rangle$  below 1K-km s<sup>-1</sup> comprise less than 20% of the total amount of CO emission.

## 4.1. The CO-H<sub>2</sub> conversion factor

Comparison of the scaled HCO<sup>+</sup> absorption and IRAM 30 emission measurements provides the most direct determination of the actual CO-H<sub>2</sub> conversion factor along the lines of sight studied in this work. Figure 7 shows the trends in  $\langle N(H_2) \rangle$ ,  $\langle W_{CO} \rangle$ , and  $\langle N(H_2) \rangle / \langle W_{CO} \rangle$  binned in 0.05 mag increments of  $E_{B-V}$ as in Figure 3. Included are the 28 Galactic anticenter sightlines where HCO<sup>+</sup> was detected, with  $W_{CO}$  taken at the  $3\sigma$  upper limit for sightlines where CO emission was not detected with greater significance. Also included is the sightline toward J1733 in Chamaeleon where CO emission was detected. The  $3\sigma$  upper limits  $W_{CO} \le 1.5 \text{ K-km s}^{-1}$  in Chamaeleon are not useful.

Figure 7 illustrates the behavior of the CO-H<sub>2</sub> conversion factor that is tabulated for different samples in Table 3. The values of  $\langle N(H_2) \rangle$  and  $\langle W_{CO} \rangle$  both increase steadily with  $E_{B-V}$ in Figure 7, but at different rates so that their ratio declines by a factor  $\approx$  7 to  $\langle N(H_2) \rangle / \langle W_{CO} \rangle = 2.5 - 3 \times 10^{20} H_2 \text{ cm}^{-2} (\text{K-}$ km s<sup>-1</sup>)<sup>-1</sup> for  $E_{B-V} \gtrsim 0.5$  mag. Variations of similar magnitude in



**Fig. 3.** Trends in mean N(HI), N(H<sub>2</sub>), and N(DNM). The results are binned in 0.05 mag intervals of  $E_{B-V}$ . Left: N(HI) is calculated over the whole line profile. Right: N(HI) = N(HI)<sub>cl</sub> the HI column density associated with the clouds studied here. The number of sightlines in each bin is shown in both panels.



Fig. 4. N(DNM) plotted against total sightline N(HI) (bottom) and cloud-associated atomic hydrogen  $N(HI)_{cl}$  (top).

individual diffuse and/or translucent MBM clouds were reported by Magnani et al. (1998) and Cotten & Magnani (2013).

Mean values of N(H<sub>2</sub>)/W<sub>CO</sub> are  $2 - 2.5 \times 10^{20}$ H<sub>2</sub> cm<sup>-2</sup>(K-km s<sup>-1</sup>)<sup>-1</sup> for the old and new samples with  $\langle W_{CO} \rangle \ge 1$  K-km s<sup>-1</sup>, increasing by factors of 2-3 for the samples with IRAM 30m detections below 1K-km s<sup>-1</sup> and upper limits. Also see the inset in Figure 7 on this point.

The values of the CO-H<sub>2</sub> conversion factor derived are comparable to those derived in extant Galactic-scale  $\gamma$ -ray analyses (Table E.1 in Remy et al. 2017) when  $W_{CO} \gtrsim 1$  K-km s<sup>-1</sup>, but are uniformly larger than those derived in cloud-level studies like the DNM analysis whose results are summarized in Table 2 of Remy et al. (2017). Those results ranged from  $1 - 1.6 \times 10^{20}$ H<sub>2</sub> cm<sup>-2</sup>(K-km s<sup>-1</sup>)<sup>-1</sup> for determinations based on dust and from  $0.44-1.00\times 10^{20} H_2~cm^{-2}~(K\mbox{-}km\,s^{-1})^{-1}$  for determinations based on gamma rays.

There is no contradiction with the larger values found in this work whose method of studying widely separated, semirandomly placed sightlines is similar to the larger scale studies. The strongly CO-emitting ( $W_{CO} > 10$  K-km s<sup>-1</sup>) regions encountered in the cloud-level studies that have generally small values of  $X_{CO}$  (see Figure 13 in Remy et al. 2018) were not sampled here.

There may also be a difference arising from the operational definition of the conversion factor. The conversion factors in Table 2 in Remy et al. (2017) are those that optimize the fit of the  $\gamma$ -ray or dust emissivity to a total hydrogen column density that is represented schematically as N(H) = N(HI)+N(DNM)+2N(H<sub>2</sub>) = N(HI)+N(DNM)+2X<sub>CO</sub>W<sub>CO</sub>. After the analysis, there remains a DNM constituent whose molecular fraction is undetermined and not explicitly considered in the definition of the multiplier  $X_{CO}$ ; we showed that the DNM is largely molecular. By contrast, the present analysis determines N(H<sub>2</sub>) independent of CO emission and defines  $X_{CO} = N(H_2)/W_{CO}$ . This N(H<sub>2</sub>) includes the molecular component of the DNM that we took pains to consider separately in the discussion of Figure 6.

# 4.2. Achievable limits and detection thresholds for CO, DNM, and $H_2$

Sightlines in our study often had rms CO emission noise  $\Delta W_{CO} \gtrsim 1/3 - 1/2$  K-km s<sup>-1</sup> in the prior DNM analysis (Table 2). For a typical Galactic conversion  $2N(H_2)/W_{CO} = 4 \times 10^{20}$  cm<sup>-2</sup> (K-kms)<sup>-1</sup>, the equivalent  $3\sigma$  threshold detection limits on the hydrogen column are  $2N(H_2) = 4 - 6 \times 10^{20}$  cm<sup>-2</sup>, above the actual values of N(DNM) along most of the sightlines we observed. By contrast, the median  $3\sigma$  detection threshold from HCO<sup>+</sup> in our work is  $2N(H_2) > 1.1 \times 10^{20}$  cm<sup>-2</sup> and typical N(DNM) values are N(DNM)  $\gtrsim 2 \times 10^{20}$  cm<sup>-2</sup> (Figure 6).

The effectiveness of reducing the detection threshold of CO emission below 1 K-km s<sup>-1</sup> can be assessed by examining the distribution of CO detections and upper limits in the IRAM 30m CO observations. The subsamples of IRAM CO observations



**Fig. 5.** N(DNM) plotted against  $E_{B-V}$  (top) and 2N(H<sub>2</sub>) (bottom). Larger symbols represent sightlines where  $W_{CO} \gtrsim 1$  K-km s<sup>-1</sup>.

summarized in Table 3 show 6 detections with  $W_{CO} > 1$  K-km s<sup>-1</sup>, 12 detections with  $W_{CO} < 1$  K-km s<sup>-1</sup>, and ten upper limits at levels  $W_{CO} < 0.1 - 0.2$  K-km s<sup>-1</sup> (see Table 1 for values of upper limits and Figure 3 for a graphical representation of the data). HCO<sup>+</sup> was detected along all of these sightlines, and the three subsamples respectively represent fractions 0.66, 0.26, and 0.08 of the total amount of H<sub>2</sub>.

Thus, a survey with a detection limit of 1 K-km s<sup>-1</sup> would detect approximately two-thirds of the molecular gas along these diffuse and/or translucent lines of sight, and a much increased effort to reduce the detection limit to 0.2 K-km s<sup>-1</sup> might find another one-fourth of the H<sub>2</sub>. Comparable reductions in the fraction of undetected H<sub>2</sub> with increasing CO sensitivity down to rms levels  $\Delta W_{CO} \approx 0.1$  K-km s<sup>-1</sup> were achieved by Donate & Magnani (2017). Missing one-third of the H<sub>2</sub> at the 1 K-km s<sup>-1</sup> threshold is consistent with the dark gas fraction derived by Wolfire et al. (2010) and Gong et al. (2018).



Fig. 6. N(DNM) plotted against  $N(H_2)$  for sightlines lacking detected CO emission in the analysis of Remy et al. (2018). Shown are loci at which the number of H nuclei in  $H_2$  is 50, 100, and 200% of that in DNM.



**Fig. 7.** Trends in mean N(H<sub>2</sub>) (black squares), W<sub>CO</sub> (pink diamonds), and N(H<sub>2</sub>)/W<sub>CO</sub> (blue triangles). The results are binned in 0.05 mag intervals of E<sub>B-V</sub>, as in Figure 3. Bins in which all sightlines have only upper limits on W<sub>CO</sub> are indicated by upper and lower limits. The scales for N(H<sub>2</sub>) cm<sup>-2</sup> and N(H<sub>2</sub>)/W<sub>CO</sub> ( cm<sup>-2</sup> (K-km s<sup>-1</sup>)<sup>-1</sup>) are read on the left vertical axis and that for W<sub>CO</sub> (K-km s<sup>-1</sup>) on the right. The number of sightlines in each bin is shown as in Figure 3 and the count is carried separately for W<sub>CO</sub> at low E<sub>B-V</sub>. The light gray dashed line is a power-law fit N(H<sub>2</sub>) =  $10^{21.0973}E_{B-V}^{1.335}$ .

#### 4.3. The wider view

In Section 4.2 we note that two-thirds of the H<sub>2</sub> was found along the sightlines with  $W_{CO} > 1 \text{ K-km s}^{-1}$  and that a deeper survey with a detection limit of 0.2 K-km s<sup>-1</sup> would have found another 25% of the H<sub>2</sub>. At this point we can ask how the results of this sparse sampling are reflected in the region as a whole. We drew a hull around the observed anticenter sightlines, as shown in Figure C.1 and derived the pixel-by-pixel statistics for the contained area, the amount of material (taken proportional to  $E_{B-V}$ ), and the amount of H<sub>2</sub>. For H<sub>2</sub> we used the N(H<sub>2</sub>)- $E_{B-V}$  relationship N(H<sub>2</sub>) =  $10^{21.0973}E_{B-V}^{1.335 \ 1}$  and the fact that  $W_{CO} \gtrsim 1 \ K\text{-km s}^{-1}$  at  $E_{B-V} \gtrsim 0.4 \ \text{mag}$  or N(H<sub>2</sub>)  $\gtrsim 4 \times 10^{20} \ \text{cm}^{-2}$  in Figure 7 (see also Figure 5 of Paper 1).

Figure 8 shows the probability densities (at left) and cumulative distributions for the derived quantities. Reading values off the cumulative probability distributions at right in Figure 8, conditions with  $E_{B-V} \geq 0.4$  mag,  $N(H_2) \gtrsim 4 \times 10^{20} \mbox{ cm}^{-2}$  occur over one-fourth of the total area containing one-half of the total projected gas column and two-thirds of the H\_2. Apparently, the sampled sightlines were representative of the region as a whole regarding the H\_2 distribution.

Sampling 90% of the H<sub>2</sub> would require detecting CO emission at  $E_{B-V} \gtrsim 0.2$  mag where  $N(H_2) \approx 2 \times 10^{20}$  cm<sup>-2</sup>. The sampling here is too sparse to derive an equivalent value of W<sub>CO</sub> in Figure 7, but the broader sample in Paper 1 suggests W<sub>CO</sub>  $\gtrsim 0.3$  K-km s<sup>-1</sup> would be appropriate. Sightlines with  $E_{B-V} < 0.32$  mag or A<sub>V</sub> < 1 mag comprise two-thirds of the area, one-third of the mass, and one-fourth of the H<sub>2</sub>.

#### 4.4. Comparison with predictions and models

For models, the critical factors in predicting the amount of dark gas are the threshold where  $W_{CO}$  reaches the 1 K-km s<sup>-1</sup> brightness level and the amount of material above the threshold. In the regime of diffuse molecular gas the integrated brightness of the J=1-0 line is determined almost exclusively by the CO column density, with  $W_{CO}$  (K-km s<sup>-1</sup>) = N(CO)/10<sup>15</sup> cm<sup>-2</sup> for hydrogen number densities n(H) = 64 – 500 cm<sup>-3</sup> (Liszt 2007, 2017; Hu et al. 2021) and kinetic temperatures appropriate to the local thermal pressure. This simple proportionality between column density and brightness persists well beyond the CO column density where the J=1-0 transition becomes optically thick, owing to the strongly subthermal excitation (Goldreich & Kwan 1974).

Thus, the CO chemistry dominates the CO brightness, and the sky map of CO emission is a map of the chemistry. The observations can be reproduced by an ad hoc CO formation chemistry in which a fixed relative abundance of HCO<sup>+</sup>  $N(HCO^+)/N(H_2) = 3 \times 10^{-9}$  thermally recombines to CO if the H<sub>2</sub> formation and shielding of H<sub>2</sub> and CO are treated selfconsistently along with the heating and cooling Liszt (2017). However, networks of chemical reactions in quiescent gas may fail to form the observed quantity of HCO<sup>+</sup> and produce too little CO. This causes the CO brightness to reach 1 K-km s<sup>-1</sup> at overly large values of E<sub>B-V</sub> and N(H<sub>2</sub>) where shielding of CO by dust and H<sub>2</sub> make up for the deficit in the CO formation rate (e.g., Gong et al. 2018 Figure 7; Hu et al. 2021 Figure 9).

Models with a weak CO formation chemistry have an innate tendency to overestimate the amount of CO-dark gas, but the actual amount of CO-dark gas depends on the distribution of material. In practice the DNM fraction seen by Remy et al. (2018) varied from 0 to 0.3 (their Figure 8) and the sightlines sampled here had  $< f_{DNM} > = 0.13$ .

#### 5. Summary

We took 89.2 GHz ALMA HCO<sup>+</sup> absorption spectra toward 33 compact millimeter-wave extragalactic continuum sources seen against the Galactic anticenter (Figure 1 and the tables above).

We also observed J=1-0 CO emission at the IRAM 30m telescope in the 28 anticenter directions where HCO<sup>+</sup> was detected. Inferring N(H<sub>2</sub>) from N(HCO<sup>+</sup>) using the ratio N(HCO<sup>+</sup>)/N(H<sub>2</sub>) =  $3 \times 10^{-9}$ , we combined these results with those from our earlier study of 13 directions where HCO<sup>+</sup> absorption was detected in the outskirts of Chamaeleon. We compared the inferred N(H<sub>2</sub>) with prior determinations of the column densities of the dark neutral medium, the neutral gas of uncertain (atomic or molecular) character that had been found to be missing in HI and/or CO emission when compared with the submillimeter dust and  $\gamma$ -ray emissivities of large-scale molecular cloud complexes.

Binning the HI, H<sub>2</sub>, and DNM column densities in reddening, we showed in Figure 3 that the mean DNM and molecular gas column densities were comparable and varied compatibly by factors of 50-100 over the observed range  $E_{B-V} = 0.09 - 1$  mag, while N(H(I) varied by only factors of 2-3. The means of N(H<sub>2</sub>) and N(DNM) are small at low mean reddening, and increase in similar fashion up to  $E_{B-V} = 0.5$  mag where molecular gas begins to dominate and CO emission is strong. N(H<sub>2</sub>) continues to increase with higher reddening, but N(DNM) and the column density of cloud-associated HI fall where H<sub>2</sub> dominates (sequestering hydrogen) and CO emission is stronger and more closely representative of N(H<sub>2</sub>).

We made detailed individual sightline-level comparisons of N(DNM) with  $E_{B-V}$ , N(HI), and  $N(H_2)$  in Figures 4-6. Sightlines with appreciable DNM appear at all N(HI) (Figure 4); the overall mean DNM fraction  $f_{DNM} = N(DNM)/(N(HI)+2N(H_2)) = 0.12$ is modest. Sightlines with appreciable DNM are lacking when  $E_{B-V} \lesssim 0.15$  mag (Figure 5) and when  $2N(H_2) \lesssim 10^{20}$  cm<sup>-2</sup> or  $2N(H_2) \gtrsim 10^{21}$  cm<sup>-2</sup> (Figure 6). In Figure 6 we compared N(DNM) and N(H<sub>2</sub>) in directions lacking detected CO emission in order to eliminate the case that H<sub>2</sub> was already represented by CO emission in the DNM analysis. This figure showed that there were 2-3 sightlines in each subsample (Chamaeleon and anticenter) or 5-6/46 overall where  $2N(H_2) \le N(DNM)/2$  and H<sub>2</sub> accounted for the minority of the DNM. There are also a few directions at  $2N(H_2) \approx 10^{21} \text{ cm}^{-2}$  where  $2N(H_2) > 2N(DNM)$ . Overall, the amounts of DNM and  $H_2$  are similar  $\langle 2N(H_2) \rangle =$ 1.3 <N(DNM)> for the unambiguous cases lacking CO emission. The form of the DNM is overwhelmingly molecular hydrogen.

Directions with  $<\!W_{CO}\!>\!>\!1$  K-km  $\rm s^{-1}$  have two times smaller mean DNM fractions  $<\!f_{DNM}\!>=0.06$ , while sightlines with  $<\!W_{CO}\!><\!1$  K-km  $\rm s^{-1}$  have three times higher  $f_{DNM}\gtrsim0.17$ -0.19 (Table 3). The relatively few sightlines with  $W_{CO}\geq1$  K-km  $\rm s^{-1}$  contain two-thirds of the  $H_2$  detected in HCO<sup>+</sup>, and detecting 90% of the  $H_2$  would require detecting CO at levels  $W_{CO}\approx0.2$ -0.3 K-km  $\rm s^{-1}$ .

The CO-H<sub>2</sub> conversion factor falls steadily with increasing  $E_{B-V}$  or  $W_{CO}$  in Figure 7. Because the H<sub>2</sub> is concentrated in the sightlines with  $W_{CO} \ge 1$  K-km s<sup>-1</sup>, the overall mean CO-H<sub>2</sub> conversion factors in our work are  $\langle N(H_2) \rangle / \langle W_{CO} \rangle = 2 - 3 \times 10^{20} H_2$  cm<sup>-2</sup> for samples of sightlines with detectable CO emission. These values are comparable to previously determined large-scale Galactic averages, and are substantially higher than the global values determined by the cloud-level analyses quoted here to derive N(DNM). We ascribed these differences in part to the present sampling of widely scattered sightlines (i.e., that we did not do a cloud-level analysis) and perhaps to differences in the operational definition of the conversion factor, as discussed in Section 4.1.

Subsequent papers derived from the observations discussed here will focus on the physics and chemistry of the molecules

 $<sup>^1</sup>$  f\_{H\_2} = 2N(H\_2)/N(H) = 0.4-0.5 at E\_{B-V} = 2 mag if N(H)/E\_{B-V} = 6 -  $8 \times 10^{21} mag^{-1}$ 



**Fig. 8.** Probability distributions of area, amount of material, and molecular hydrogen with respect to  $E_{B-V}$ . Left: Counts (probability density) scaled to the same area. Right: Cumulative distribution function. N(H<sub>2</sub>) is calculated using the mean power-law fit N(H<sub>2</sub>) =  $10^{21.0973}E_{B-V}^{1.335}$  derived as shown in Figure 7.

| Source     | RA(J2000) | Dec(J2000) | l        | b        | $E_{B-V}^{1}$ | 89 GHz flux | $\Upsilon_{HCO}^{+2}$ | $\sigma \Upsilon_{\rm HCO^+}{}^3$ | $W_{CO}^4$           | $\sigma W_{CO}{}^5$ |
|------------|-----------|------------|----------|----------|---------------|-------------|-----------------------|-----------------------------------|----------------------|---------------------|
|            | hh.mmsss  | dd.mmsss   | degrees  | degrees  | mag           | Jy          | $km s^{-1}$           | $km s^{-1}$                       | K-km s <sup>-1</sup> | $K-km s^{-1}$       |
| J0203+1134 | 2.03464   | 11.34492   | 149.6826 | -47.4992 | 0.144         | 0.126       | ≤0.263                | 0.088                             |                      |                     |
| J0209+1352 | 2.09357   | 13.52045   | 150.1800 | -44.8290 | 0.094         | 0.223       | 0.20                  | 0.050                             | ≤0.071               | 0.024               |
| J0211+1051 | 2.11132   | 10.51348   | 152.5781 | -47.3674 | 0.145         | 0.462       | 0.76                  | 0.029                             | 0.36                 | 0.025               |
| J0213+1820 | 2.13105   | 18.20255   | 148.7289 | -40.4014 | 0.130         | 0.093       | ≤0.345                | 0.115                             |                      |                     |
| J0231+1322 | 2.31459   | 13.22547   | 157.0917 | -42.7380 | 0.121         | 0.430       | 0.14                  | 0.025                             | ≤0.064               | 0.021               |
| J0242+1742 | 2.42243   | 17.42589   | 157.0180 | -37.7033 | 0.077         | 0.227       | ≤0.151                | 0.050                             |                      |                     |
| J0252+1718 | 2.52077   | 17.18427   | 159.7420 | -36.7885 | 0.220         | 0.172       | 0.25                  | 0.077                             | ≤0.096               | 0.032               |
| J0325+2224 | 3.25368   | 22.24004   | 163.6700 | -28.0213 | 0.213         | 1.162       | 1.01                  | 0.017                             | 0.94                 | 0.051               |
| J0329+3510 | 3.29154   | 35.10060   | 155.9152 | -17.4042 | 0.267         | 0.570       | 0.51                  | 0.032                             | ≤0.143               | 0.048               |
| J0329+2756 | 3.29577   | 27.56155   | 160.7030 | -23.0743 | 0.198         | 0.158       | ≤0.193                | 0.064                             |                      |                     |
| J0334+0800 | 3.34533   | 8.00144    | 177.2396 | -37.0871 | 0.391         | 0.150       | 0.44                  | 0.088                             | ≤0.162               | 0.054               |
| J0336+3218 | 3.36301   | 32.18293   | 158.9998 | -18.7650 | 0.733         | 1.689       | 0.16                  | 0.009                             | ≤0.219               | 0.073               |
| J0356+2903 | 3.56085   | 29.03423   | 164.6120 | -18.4927 | 0.212         | 0.139       | 1.50                  | 0.090                             | 1.62                 | 0.042               |
| J0357+2319 | 3.57216   | 23.19538   | 169.0302 | -22.4661 | 0.185         | 0.224       | 0.28                  | 0.027                             | ≤0.192               | 0.064               |
| J0400+0550 | 4.00117   | 5.50431    | 184.2710 | -33.7266 | 0.266         | 0.159       | 0.25                  | 0.063                             | ≤0.172               | 0.057               |
| J0401+0413 | 4.01199   | 4.13344    | 186.0261 | -34.4947 | 0.341         | 0.405       | 0.49                  | 0.021                             | 0.19                 | 0.048               |
| J0403+2600 | 4.03056   | 26.00015   | 168.0260 | -19.6483 | 0.201         | 0.331       | 0.60                  | 0.029                             | 0.62                 | 0.067               |
| J0406+0637 | 4.06343   | 6.37150    | 184.7075 | -32.0009 | 0.283         | 0.264       | 0.54                  | 0.051                             | 0.63                 | 0.040               |
| J0407+0742 | 4.07291   | 7.42075    | 183.8723 | -31.1558 | 0.265         | 0.387       | 0.53                  | 0.031                             | 0.19                 | 0.042               |
| J0426+0518 | 4.26366   | 5.18199    | 189.3631 | -28.7705 | 0.291         | 0.516       | 0.12                  | 0.020                             | ≤0.170               | 0.057               |
| J0426+2327 | 4.26557   | 23.27396   | 173.8881 | -17.4457 | 0.539         | 0.304       | 2.57                  | 0.057                             | 4.84                 | 0.045               |
| J0427+0457 | 4.27476   | 4.57083    | 189.8857 | -28.7306 | 0.335         | 0.414       | 0.62                  | 0.024                             | 0.39                 | 0.045               |
| J0437+2037 | 4.31038   | 20.37343   | 176.8096 | -18.5565 | 0.532         | 0.217       | 1.54                  | 0.073                             | 0.67                 | 0.026               |
| J0431+1731 | 4.31574   | 17.31358   | 179.4942 | -20.3579 | 0.464         | 0.104       | 1.01                  | 0.110                             | 0.70                 | 0.049               |
| J0433+0521 | 4.33111   | 5.21156    | 190.3730 | -27.3967 | 0.298         | 4.911       | 0.35                  | 0.003                             | ≤0.109               | 0.036               |
| J0437+2940 | 4.37044   | 29.40138   | 170.5818 | -11.6609 | 0.979         | 0.059       | 5.92                  | 1.138                             | 10.42                | 0.027               |
| J0438+3004 | 4.38049   | 30.04455   | 170.4116 | -11.2283 | 0.952         | 0.689       | 6.25                  | 0.038                             | 7.11                 | 0.026               |
| J0439+3045 | 4.39178   | 30.45076   | 170.0655 | -10.5913 | 0.867         | 0.195       | 5.05                  | 0.082                             | 6.69                 | 0.027               |
| J0440+1437 | 4.40211   | 14.37570   | 183.2538 | -20.5438 | 0.681         | 0.337       | 1.21                  | 0.031                             | 0.83                 | 0.029               |
| J0445+0715 | 4.45014   | 7.15539    | 190.4535 | -23.8898 | 0.121         | 0.275       | ≤0.083                | 0.028                             |                      |                     |
| J0449+1121 | 4.49077   | 11.21286   | 187.4274 | -20.7365 | 0.504         | 0.521       | 0.65                  | 0.022                             | 0.23                 | 0.033               |
| J0502+1338 | 5.02332   | 13.38110   | 187.4143 | -16.7456 | 0.564         | 0.271       | 1.81                  | 0.059                             | 2.46                 | 0.031               |
| J0510+1800 | 5.10024   | 18.00416   | 184.7304 | -12.7895 | 0.328         | 2.411       | 0.13                  | 0.005                             | 0.17                 | 0.036               |

<sup>1</sup>From Schlegel et al. (1998)

<sup>2</sup>Integrated optical depth or  $3\sigma$  upper limit over a 3 km s<sup>-1</sup> interval

<sup>3</sup>Integrated emission from our results or  $3\sigma$  upper limit over a 3 km s<sup>-1</sup> interval

<sup>4</sup> rms of detected emission or rms over a 3km s<sup>-1</sup> interval

Table 2. Line-of-sight properties and derived quantities

| Source     | E(B-V) | N(HI) <sup>1</sup>        | N(HI) <sub>cl</sub> <sup>1</sup> | $2N(H_2)^2$               | $W_{CO}^{3}$         | N(DNM) <sup>1</sup>       | 2N(H <sub>2</sub> )/N(HI) N(DNM)/N(H |       | $X_{\rm CO}^4$ |
|------------|--------|---------------------------|----------------------------------|---------------------------|----------------------|---------------------------|--------------------------------------|-------|----------------|
|            | mag    | $10^{21} \text{ cm}^{-2}$ | $10^{21} \text{ cm}^{-2}$        | $10^{21} \text{ cm}^{-2}$ | K-km s <sup>-1</sup> | $10^{21} \text{ cm}^{-2}$ |                                      |       |                |
| J0203+1134 | 0.144  | 0.664                     | 0.646                            | ≤0.193                    |                      | 0.201                     | ≤0.290                               | 0.303 |                |
| J0209+1352 | 0.094  | 0.696                     | 0.686                            | 0.144                     |                      | 0.00                      | 0.206                                | 0.00  |                |
| J0211+1051 | 0.145  | 0.618                     | 0.617                            | 0.561                     |                      | 0.240                     | 0.908                                | 0.388 |                |
| J0213+1820 | 0.130  | 0.933                     | 0.911                            | ≤0.253                    |                      | 0.177                     | ≤0.271                               | 0.190 |                |
| J0231+1322 | 0.121  | 0.926                     | 0.852                            | 0.103                     |                      | 0.00                      | 0.111                                | 0.00  |                |
| J0242+1742 | 0.077  | 0.820                     | 0.819                            | ≤0.111                    |                      | 0.00                      | ≤0.135                               | 0.00  |                |
| J0252+1718 | 0.220  | 1.142                     | 1.139                            | 0.184                     |                      | 0.299                     | 0.161                                | 0.262 |                |
| J0325+2224 | 0.213  | 0.999                     | 0.926                            | 0.742                     | 2.82                 | 0.180                     | 0.743                                | 0.180 | 1.314          |
| J0329+3510 | 0.267  | 1.418                     | 1.131                            | 0.372                     |                      | 0.150                     | 0.263                                | 0.106 |                |
| J0329+2756 | 0.198  | 1.129                     | 1.002                            | ≤0.142                    | $\leq 1.97$          | 0.00                      | ≤0.126                               | 0.00  |                |
| J0334+0800 | 0.391  | 1.893                     | 1.855                            | 0.322                     |                      | 0.223                     | 0.170                                | 0.118 |                |
| J0336+3218 | 0.733  | 1.435                     | 1.165                            | 0.119                     | ≤2.60                | 0.606                     | 0.082                                | 0.423 |                |
| J0356+2903 | 0.212  | 0.666                     | 0.520                            | 1.099                     | 1.40                 | 0.178                     | 1.650                                | 0.267 | 3.931          |
| J0357+2319 | 0.185  | 1.022                     | 0.944                            | 0.205                     | ≤0.79                | 0.00                      | 0.200                                | 0.00  |                |
| J0400+0550 | 0.266  | 1.266                     | 1.210                            | 0.182                     |                      | 0.238                     | 0.144                                | 0.188 |                |
| J0401+0413 | 0.341  | 1.209                     | 1.202                            | 0.357                     |                      | 0.426                     | 0.296                                | 0.352 |                |
| J0403+2600 | 0.201  | 0.830                     | 0.667                            | 0.438                     | 1.99                 | 0.00                      | 0.528                                | 0.00  | 1.101          |
| J0406+0637 | 0.283  | 1.182                     | 1.143                            | 0.397                     |                      | 0.266                     | 0.335                                | 0.225 |                |
| J0407+0742 | 0.265  | 1.033                     | 1.006                            | 0.387                     |                      | 0.258                     | 0.374                                | 0.250 |                |
| J0426+0518 | 0.291  | 1.305                     | 1.141                            | 0.091                     | ≤1.40                | 0.414                     | 0.070                                | 0.317 |                |
| J0426+2327 | 0.539  | 1.586                     | 1.331                            | 1.887                     | 5.78                 | 0.00                      | 1.190                                | 0.00  | 1.632          |
| J0427+0457 | 0.335  | 1.361                     | 1.192                            | 0.457                     | 2.49                 | 0.590                     | 0.336                                | 0.433 | 0.918          |
| J0437+2037 | 0.532  | 2.324                     | 1.992                            | 1.130                     | ≤1.61                | 0.00                      | 0.486                                | 0.00  |                |
| J0431+1731 | 0.464  | 1.714                     | 1.457                            | 0.739                     | 3.09                 | 0.00                      | 0.431                                | 0.00  | 1.198          |
| J0433+0521 | 0.298  | 1.159                     | 0.950                            | 0.255                     | ≤1.48                | 0.795                     | 0.220                                | 0.686 |                |
| J0437+2940 | 0.979  | 1.320                     | 0.936                            | 4.345                     | 11.98                | 0.126                     | 3.292                                | 0.095 | 1.814          |
| J0438+3004 | 0.952  | 2.019                     | 1.002                            | 4.586                     | 7.70                 | 0.771                     | 2.271                                | 0.382 | 2.979          |
| J0439+3045 | 0.867  | 2.361                     | 0.745                            | 3.701                     | 9.42                 | 0.00                      | 1.567                                | 0.00  | 1.964          |
| J0440+1437 | 0.681  | 1.659                     | 1.524                            | 0.888                     | ≤1.97                | 1.230                     | 0.535                                | 0.741 |                |
| J0445+0715 | 0.121  | 1.044                     | 0.915                            | ≤0.061                    |                      | 0.00                      | ≤0.058                               | 0.00  |                |
| J0449+1121 | 0.504  | 1.416                     | 1.347                            | 0.476                     |                      | 0.791                     | 0.336                                | 0.558 |                |
| J0502+1338 | 0.564  | 2.188                     | 1.957                            | 1.324                     | ≤1.90                | 0.672                     | 0.605                                | 0.307 |                |
| J0510+1800 | 0.328  | 2.347                     | 1.986                            | 0.092                     | ≤2.00                | 0.00                      | 0.039                                | 0.00  |                |
| J0942-7731 | 0.336  | 1.000                     | 0.617                            | 0.837                     | ≤1.50                | 0.382                     | 0.838                                | 0.382 |                |
| J1058-8003 | 0.152  | 0.651                     | 0.470                            | 0.147                     | "                    | 0.371                     | 0.226                                | 0.570 |                |
| J1136-6827 | 0.460  | 2.407                     | 0.915                            | 0.910                     | "                    | 0.172                     | 0.378                                | 0.071 |                |
| J1145-6954 | 0.387  | 1.925                     | 0.998                            | 0.638                     | "                    | 0.223                     | 0.331                                | 0.116 |                |
| J1147-7935 | 0.300  | 2.506                     | 0.773                            | 0.040                     | "                    | 0.00                      | 0.016                                | 0.00  |                |
| J1152-8344 | 0.283  | 1.088                     | 0.505                            | 0.176                     | "                    | 0.515                     | 0.162                                | 0.473 |                |
| J1224-8313 | 0.257  | 0.962                     | 0.563                            | 0.693                     | "                    | 0.335                     | 0.720                                | 0.348 |                |
| J1254-7138 | 0.282  | 1.902                     | 0.660                            | 0.102                     | **                   | 0.00                      | 0.053                                | 0.00  |                |
| J1312-7724 | 0.476  | 1.257                     | 0.671                            | 0.199                     | "                    | 0.396                     | 0.158 0.315                          |       |                |
| J1550-8258 | 0.107  | 0.679                     | 0.416                            | 0.177                     | "                    | 0.027                     | 0.261                                | 0.039 |                |
| J1617-7717 | 0.091  | 0.659                     | 0.431                            | 0.043                     | "                    | 0.045                     | 0.065                                | 0.068 |                |
| J1723-7713 | 0.255  | 0.834                     | 0.500                            | 1.105                     | 2.40                 | 0.195                     | 1.325                                | 0.234 | 2.302          |
| J1733-7935 | 0.139  | 0.792                     | 0.525                            | 0.041                     | $\leq 1.50$          | 0.00                      | 0.052                                | 0.00  |                |

<sup>1</sup> HI and DNM column densities are from Planck Collaboration et al. (2015) and Remy et al. (2017, 2018)

 $^{2}$  N(H<sub>2</sub>) = N(HCO<sup>+</sup>)/3 × 10<sup>-9</sup>

<sup>3</sup> Integrated J=1-0 CO emission used by Planck Collaboration et al. (2015) and Remy et al. (2017, 2018)

 $^{4} X_{\rm CO} = {\rm N(H_2)}/{\rm W_{\rm CO}}$ 

observed in the course of this work, chiefly HCO<sup>+</sup>, C<sub>2</sub>H, and HCN.

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| Table 3. Mean | line-of-sight | properties an | nd derived | quantities |
|---------------|---------------|---------------|------------|------------|
|---------------|---------------|---------------|------------|------------|

| Sample   | E(B-V) | N(HI)                     | N(HI) <sub>cl</sub>       | $2N(H_2)$                 | W <sub>CO</sub>      | N(DNM)                    | $f_{H_2}$ | f <sub>DNM</sub> | X <sub>CO</sub> |
|--|--------|---------------------------|---------------------------|---------------------------|----------------------|---------------------------|-----------|------------------|-----------------|
|  | mag    | $10^{21} \text{ cm}^{-2}$ | $10^{21} \text{ cm}^{-2}$ | $10^{21} \text{ cm}^{-2}$ | K-km s <sup>-1</sup> | $10^{21} \text{ cm}^{-2}$ | 2         |                  |                 |
| Galactic anticenter                                  | 0.362  | 1.324                     | 1.119                     | 0.798                     | 5.185                | 0.268                     | 0.38      | 0.13             | 1.93            |
| #  | 33     | 33                        | 33                        | 33                        | 9                    | 33                        | 33        | 33               | 9               |
| Chamaeleon   | 0.271  | 1.282                     | 0.619                     | 0.393                     | 2.400                | 0.205                     | 0.23      | 0.12             | 2.30            |
| #  | 13     | 13                        | 13                        | 13                        | 1                    | 13                        | 13        | 13               | 1               |
| All LOS  | 0.336  | 1.312                     | 0.977                     | 0.684                     | 4.906                | 0.250                     | 0.34      | 0.13             | 1.95            |
| #  | 46     | 46                        | 46                        | 46                        | 10                   | 46                        | 46        | 46               | 10              |
| $E_{B-V} \le 0.2mag$                                 | 0.131  | 0.818                     | 0.710                     | 0.168                     |                      | 0.082                     | 0.18      | 0.08             |                 |
| #  | 13     | 13                        | 13                        | 13                        |                      | 13                        | 13        | 13               |                 |
| Lacking HCO <sup>+1</sup>                            | 0.134  | 0.918                     | 0.859                     | ≤0.152                    |                      | 0.076                     | ≤0.14     | ≥0.08            |                 |
| #  | 5      | 5                         | 5                         | 5                         |                      | 5                         | 5         | 5                |                 |
| $DNM \le 10^{20} \text{ cm}^{-2}$                    | 0.273  | 1.373                     | 0.953                     | 0.539                     | 5.070                | 0.004                     | 0.28      | 0.00             | 1.67            |
| #  | 17     | 17                        | 17                        | 17                        | 4                    | 17                        | 17        | 17               | 4               |
| CO detected earlier <sup>2</sup>                     | 0.502  | 1.369                     | 0.928                     | 1.910                     | 4.906                | 0.204                     | 0.58      | 0.06             | 1.95            |
| #  | 10     | 10                        | 10                        | 10                        | 10                   | 10                        | 10        | 10               | 10              |
| 30m CO detected                                      | 0.467  | 1.491                     | 1.197                     | 1.312                     | 2.170                | 0.318                     | 0.47      | 0.11             | 3.02            |
| #  | 18     | 18                        | 18                        | 18                        | 18                   | 18                        | 18        | 18               | 18              |
| $30 \text{m W}_{\text{CO}} > 1 \text{K-km s}^{-1}$   | 0.685  | 1.690                     | 1.082                     | 2.824                     | 5.522                | 0.291                     | 0.63      | 0.06             | 2.56            |
| #  | 6      | 6                         | 6                         | 6                         | 6                    | 6                         | 6         | 6                | 6               |
| $30 \text{m W}_{\text{CO}} \le 1 \text{K-km s}^{-1}$ | 0.358  | 1.391                     | 1.255                     | 0.555                     | 0.494                | 0.332                     | 0.29      | 0.17             | 5.62            |
| #  | 12     | 12                        | 12                        | 12                        | 12                   | 12                        | 12        | 12               | 12              |
| $30m UL \leq 1K \cdot km s^{-1}$                     | 0.287  | 1.226                     | 1.107                     | 0.198                     | < 0.140              | 0.273                     | 0.14      | 0.19             | >7.07           |
| #  | 10     | 10                        | 10                        | 10                        | 10                   | 10                        | 10        | 10               | 10              |
| Sample   | E(B-V) | N(HI)                     | N(HI) <sub>cl</sub>       | $2N(H_2)$                 | W <sub>CO</sub>      | N(DNM)                    | $f_{H_2}$ | f <sub>DNM</sub> | X <sub>CO</sub> |
|  | mag    | $10^{21} \text{ cm}^{-2}$ | $10^{21} \text{ cm}^{-2}$ | $10^{21} \text{ cm}^{-2}$ |                      | $10^{21} \text{ cm}^{-2}$ | -         |                  |                 |

<sup>1</sup> Limits in this table use  $3\sigma$  upper limits on undetected quantities

 $^2$  All sightlines detected earlier in CO had  $W_{\rm CO}$  >1K-km  $s^{-1}$ 

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Fig. A.1. Integrated HCO<sup>+</sup> optical depth  $\Upsilon_{\text{HCO}^+}$  plotted against its rms error. The upper limits along five sightlines not detected in HCO<sup>+</sup> are  $3\sigma$ .

## Appendix A: Statistics of HCO<sup>+</sup> detections

HCO<sup>+</sup> absorption was not detected in five directions, all in the Galactic anticenter sample. Figure A.1 shows  $\Upsilon_{\text{HCO}^+}$  plotted against its rms error  $\Delta\Upsilon_{\text{HCO}^+}$ , in essence against the strength of the continuum background since all sightlines were observed for the same length of time. The plot shows that even the noisiest sightlines miss relatively small amounts of molecular gas. The bias in the plot, whereby  $<\Upsilon_{\text{HCO}^+}>$  increases with  $\Delta\Upsilon_{\text{HCO}^+}$ , was to some extent built into the observing to increase the yield. That is, the source selection began with flux-limited samples that would achieve a minimum signal-to-noise ratio on the weakest source, and added 50% additional targets at lower flux  $S_{\nu}$  and higher  $E_{\text{B}-\text{V}}/\text{S}_{\nu}$  that could be serendipitously observed without a proportionate increase in the required observing time.

## Appendix B: Spectra of HCO<sup>+</sup> and CO

Spectra of HCO<sup>+</sup> absorption and CO emission in the 28 directions with detected HCO<sup>+</sup> absorption are shown in Figure B.1. The HCO<sup>+</sup> absorption is the negative-going signal shown in red. The black histogram shows the CO emission. The frequencyswitched IRAM 30m CO spectra were delivered only after folding in frequency, preventing a separation of the phases of the frequency-switching cycle using the methods of Liszt (1997). Negative-going features in the CO spectrum are artifacts; only the positive-going CO signal coincident with HCO<sup>+</sup> absorption is interstellar.

## Appendix C: Area used to derive large-scale statistics

To derive the statistical distributions shown in Figure 8 and discussed in Section 4.4, we drew a hull around the observed sightlines as illustrated in Figure C.1 and summed over the interior pixels.



**Fig. B.1.** Spectra of HCO<sup>+</sup> absorption (red) and CO emission (black) from the Galactic anticenter sample. For an explanation of the appearance of the frequency-switched IRAM 30m emission spectra, see Appendix B.



**Fig. C.1.** As in Figure 1, left, but showing the outlines of an area containing the observed sightlines used to derive large-scale properties of the anticenter region (see Section 4.3).